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Physics. — *The Specific Heats of Solid Substances at the Temperatures Attainable with the Help of Liquid Helium. I. Measurements of the Atomic Heat of Lead.* By Prof. W. H. KEESOM and Dr. DONALD H. ANDREWS [1]). (Communication No. 185a from the Physical Laboratory, Leiden.

§ 1. *Introduction.* In the range of temperatures which can be reached with the help of liquid helium there have been scarcely any measurements of the specific heats of solids. KAMERLINGH ONNES and HOLST [2]) published a few preliminary measurements of the specific heat of mercury, which were made in order to find out whether a discontinuous change in the specific heat occurs at the temperature at which mercury becomes super-conductive. KAMERLINGH ONNES and HOLST concluded from these measurements, which however were not very exact, that no such discontinuity occurs.

In the present communication there are discussed some measurements of the specific heat of lead between the temperatures 2° K. and 20° K.

We selected lead as the initial substance for the study of specific heat at extremely low temperatures, primarily because lead belongs to those elements, whose specific heat falls off least rapidly with the lowering of the temperature, thus enhancing the accuracy of observation; besides this it seemed desirable because some exact observations [3]) of the specific heat of lead had been made in the region of temperature just above the range which we planned to investigate.

Particularly at the lowest temperatures, our measurements do not have the accuracy which we hoped they would and we plan to repeat them and continue them; but in the meantime by the departure of one of us it seems desirable to publish the results which we have already secured.

§ 2. *Method.* We employed the method of NERNST and EUCKEN for determining specific heat, in essentials in the way already followed by

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KEESOM and KAMERLINGH ONNES [1]) for determining specific heats at the temperatures which can be reached with the help of liquid hydrogen.

For these experiments there was constructed a combination unit for measuring temperature and for heating (see Fig. 2, Comm. N⁰.143), consisting of two concentric cylinders of copper, the outer being threaded on the outside. On the inner cylinder there was wound bifilarly a coil of insulated constantan wire, and the remaining space between the two cylinders was filled with amalgam as described in Comm. No. 143.

This constantan coil served for the heating as well as for measuring temperature. It consisted of three parts, one of one ohm, one of one hundred ohms and one of ten ohms. The wires leading to the coil were so arranged that any one of these parts could be used at will. In these experiments only the 100 ohm coil was used.

The measurements of resistance for determining the course of temperature before and after heating were made with the help of a potentiometer. During the heating this was disconnected.

The heat imparted was calculated from the mean resistance of the heating coil, from the current strength as read on a milliammeter, and from the time as adjusted by the switching device described in Comm. No. 143, Fig. 7. The switch was actually controlled by an electric impulse sent out by the clock as was the case in the measurements of Comm. No. 143. For a control the time was read from a watch.

The current used for measuring temperature was about 0.5 mA. and that for heating from 10 to 50 mA. The heat from the measuring current was taken into consideration as was necessary.

The vacuum within the calorimeter jacket was secured and maintained by a three stage diffusion pump from LEYBOLD.

§ 3. *Accuracy of the Measurements*. *a*. A factor of uncertainty is introduced in that the resistance of the constantan heating coil during heating is taken to be the mean of the resistances before and after heating. Since there is but a very small change in the resistance of the constantan for a rise even of ten degrees, the error due to the temporary rise in temperature during heating cannot exceed some few percents.

The temperature-time curve gave no reason for concluding that the temperature of the wire during heating rose very high above that of the surroundings, moreover our results at higher temperatures are in accord with those of KEESOM and KAMERLINGH ONNES, indicating that the above assumption is permissible within the calculated limit of error.

Considering that at lower temperatures much smaller currents were used for heating, it does not appear likely that this sort of error could have been greater in that region.

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[1]) These Proc. **17**, 894, 1914—1915; Comm. Leiden N⁰. 143. Also These Proc. **18**, 484, 1915—1916; Comm. Leiden N⁰. 147*a*.

b. The apparent decrease in the resistance immediately after heating at the lowest temperatures (see § 5) makes the measurements below 4° K. somewhat uncertain. Above 4° K. this cause of uncertainty does not appear to be significant.

c. The measurements of the heat capacity of the core are less accurate apparently because its heat capacity is so small. For just this reason however, the core has a relatively unimportant part of the total heat capacity in the measurements of the specific heat of lead.

d. A number of measurements is uncertain because the course of the temperature after heating was not observed over a long enough period to permit an accurate calculation of the temperature rise following the heating.

e. The following table gives an estimate of the order of magnitude of the different sources of error expressed in percent of the atomic heat capacity of lead.

Factor	2 — 4° K.	4 — 8° K.	8 — 16° K.
1. Weight of the lead block	0.05 %	0.05 %	0.05 %
2. Temperature rise	10.0	3.0	2.0
3. Energy imparted			
(current)^2	1.0	1.0	1.0
Resistance	1.0	1.0	3.0
Time	0.1	0.1	0.1
4. Part of the heat capacity due to the core	7.0	4.0	0.1
Total	19 %	9 %	6 %
Average deviation of the results from the mean	20 %	7 %	2 %
Uncertainty in the heat capacity of the core	50 %	20 %	3 %

For some measurements the deviation is considerably greater without there being any evident cause.

§ 4. *The resistance thermometer.* The data for the calibration of the constantan resistance thermometer are given in Tables I*a* and I*b*.

The temperature of the liquid helium was read off a graph based on

the formula for the vapor pressure by KAMERLINGH ONNES and WEBER [1]), excepting the lowest three temperatures which were calculated from the formula of VERSCHAFFELT, given in Comm. Leiden, N⁰. 181, p. 18, note 1 [2]).

TABLE Ia.			
Calibration of constantan thermometer in liquid helium			
Date	Vapor pressure of He mm. Hg	Temperature °K.	Resistance Ohms
20 November 1925	874.5	4.351	90.4244?
	845.1	4.313	.4233
	822.1	4.283	.4205
	784.5	4.233	.4136
	751.0	4.188	.4095
	744.8	4.180	.4088
	736.1	4.168	.4110?
	614.9	3.996	.3877
	393.5	3.599	.3425
	166.9	3.008	.2769
	59.8	2.481	.2219
	29.9	2.192	.1947
	5.21	1.578	.1522
	3.63	1.448	.1482
	3.44	1.436	.1478
17 March 1926	767.6	4.210	.4121 [3]
	767.6	4.210	.4128

Between liquid helium and liquid hydrogen temperatures interpolation was made with the help of a graph.

[1]) These Proc. 18, 493, 1915—1916; Comm. Leiden N⁰. 147b § 7.

[2]) The newer formula of VERSCHAFFELT, Comm. Leiden Suppl. N⁰. 49, p. 26, gives temperatures, which are 0.06° higher.

[3]) During the hour preceding this measurement there was no current passing through the constantan wire; for the fifteen minutes just before the next measurement there was a current of 1 milliampere. This explains the somewhat higher value of the resistance according to the second measurement, as the result of the slightly higher temperature in the thermometer core due to the heating effect of the current.

TABLE 1b.

Calibration Constantan Thermometer in liquid hydrogen

Date	Vapor Pressure H_2 mm. Hg	Temperature °K.	Resistance Ohms
5 November 1925	—	14.112 *)	91.6667
22 April 1926	754.9	20.338 **)	92.4507
„ „ „	60.0	14.115 **)	91.6678

*) Secured from simultaneous readings with Platinum thermometers Pt_{32} and Pt_{36}.
**) Secured from the vapor pressure of hydrogen according to Comm. Leiden N⁰. 156b [1]).

§ 5. *The Heat Capacity of the Heater-Thermometer-Core.*

Table II gives the data secured from the measurements of the heat capacity of the heater-thermometer-core.

A number of observations, not included in the table, did not give good results. For several of these, especially at the lowest temperatures, the energy added was too small to give a good measurable temperature rise. In some cases the behaviour of the galvanometer was as if the temperature was actually lowered by heating. The cause of this irregularity is as yet unknown to us.

For a number of measurements shown in the table, the deduction of the corrected final temperature after heating from the different successive readings of the galvanometer was uncertain. These measurements are marked with an *. In this connection it should be noted that, after the current for heating has passed, the heating wire has a temperature appreciably higher than that of the surrounding metal. There is in most cases about ten minutes before the temperature of this wire, serving also as thermometer, is the same as the temperature of the block of metal. Only when the temperature of the bath (in which it was impossible to use a stirrer [2])) remained sufficiently constant over a time long enough to provide a regular change in temperature for the block, was it possible to get the final temperature reading for the calorimetric calculation with sufficient accuracy.

These remarks apply also to the measurements of the heat capacity of the lead block communicated in § 6.

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[1]) J. PALACIOS MARTINEZ and H. KAMERLINGH ONNES. Arch. Néerl. (3A) **6**, 31, 1922. Comm. Leiden N⁰, 156b.

[2]) To promote uniformity of temperature in the bath, the calorimeter vessel was encased in a copper cylinder, cp. These Proc. **20**, p. 1000; Comm. Leiden, N⁰. 153a, § 1.

TABLE II.							
Date N°.	Current mA.	Mean Resistance Ohms	Time sec.	Temperature rise degrees K.	Mean Temperature °K.	Heat Capacity Cal./degr. K.	Heat Capacity per mol lead in the block
21 May 1926							
A XIV	18.82	90.29	36	1.98	3.07	0.1389	0.0411
XV	7.66	90.62	30	0.96	5.94	0.0398	118*
XVI	7.66	90.71	30	0.72	6.66	0.053	163*
B XII	7.67	90.33	90	1.86	3.43	0.0616	182*
XIII	7.66	90.57	30	0.34	5.58	0.1122	[332]
XIV	7.66	90.62	30	0.29	5.94	0.1316	391*
XV	7.66	90.70	60	0.81	6.56	0.0943	278*
XVI	7.65	90.90	180	2.61	8.14	0.0879	261*
XVII	7.65	91.26	300	4.26	10.99	0.0901	266
XVIII	7.65	91.63	300	3.51	13.79	0.1098	357
XIX	7.65	91.91	300	2.68	16.03	0.1440	426
XX	7.65	92.20	481	3.12	18.37	0.1989	592
XXI	7.64	92.33	599	2.99	20.95	0.2588	767
2 June 1926							
C XVIII	7.69	90.29	60	0.23	3.13	0.3081	912*
XX	7.52	90.67	180	2.93	6.30	0.0749	222*
XXI	7.50	90.91	90	1.71	8.22	0.0642	190*
D III	7.51	90.20	120	0.45	2.29	0.3231	957**
VI	7.50	90.65	90	1.21	6.18	0.0887	263*
VII	7.49	90.82	90	1.99	7.87	0.0550	163*
VIII	7.49	90.49	180	1.65	4.87	0.1322	391*

For the bracketed readings in Tables II and III there were other reasons for uncertainty, e. g. that for the measurement the temperature rise was not sufficiently large.

The results are shown in Fig. 1. The points within broken circles are those indicated in Table II as less accurate. For the calculation of the specific heat of lead in § 6 we used the line drawn through the points, (shown in the uncertain region as a dotted line).

§ 6. *The Atomic Heat of Lead.* The results of the measurements are given in Table III. The lead block [1]) weighed 699.73 grams.
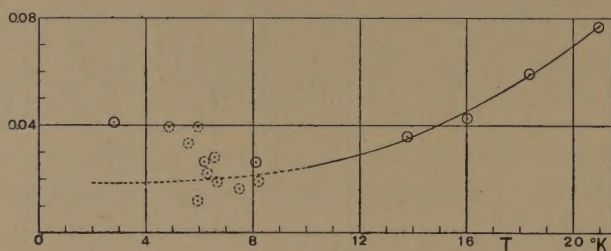
Fig. 1.
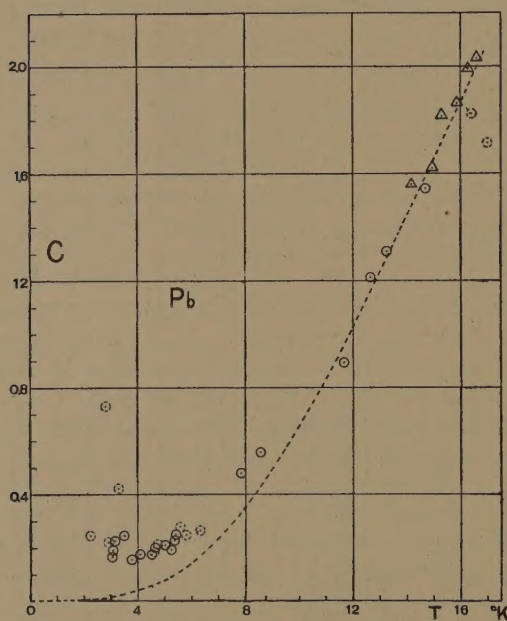
Fig. 2.

For remarks on the order of certainty of the results cf. § 5.

The results are shown graphically in Fig. 2 (dotted circles indicating less accuracy).

The △ △ mark the points secured from the accurate measurements of

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[1]) The same block as that which served for the measurements of these Proc. **17**, 894, 1914—1915; Comm. Leiden N⁰. 143, diminished by a strip which served for the resistance measurements given in these Proc. **18**, 487, note 3, 1915—1916; Comm. Leiden N⁰. 147a, p. 6, note 3.

Date N°.	Current mA.	Mean Resistance Ohms	Time sec.	Temperature-rise degrees K.	Mean Temperature °K.	Heat Capacity of core per mol lead	Atomic heat lead[1])
30 March 1926			In bath of liquid helium.				
II	27.03	90.86	64	0.600	7.84	0.021	0.480
III	27.02	90.96	120	0.975	8.57	0.022	0.557
IV	26.92	91.48	240	0.907	12.66	0.031	1.211
V	26.92	91.96	360	0.908	16.40	0.047	1.823*
VI	16.47	90.27	60	0.139	2.81	0.018	[0.728]
VII	27.20	90.33	30	0.540	3.485	0.018	0.244
VIII	27.29	90.46	60	1.41	4.61	0.019	0.183*
IX	27.17	90.67	120	2.00	6.33	0.020	0.264*
22 April			In bath of solid or liquid hydrogen				
I	47.33	91.36	60	0.94	11.66	0.028	0.893
II	55.60	91.56	68	1.03	13.26	0.033	1.308
III	60.76	91.69	120	1.82	14.69	0.038	1.543
IV	varied	91.98	239	2.79	17.00	0.050	[1.713]
V	,,	92.32	180	1.81	19.30	0.065	[2.300]
30 April			In bath of liquid helium.				
III	23.81	90.25	60	0.916	2.91	0.018	0.219*
IV	24.08	90.36	30	0.613	3.79	0.019	0.157
V	23.74	90.46	45	0.742	4.64	0.019	0.200
VI	23.75	90.55	60	0.81	5.41	0.019	0.249
IX	35.37	90.29	60	2.302	3.07	0.018	0.190
XI	23.82	90.29	120	2.355	3.04	0.018	0.166
XII	23.60	90.45	30	0.556	4.52	0.019	0.174
XIII	23.60	90.50	30	0.47	5.01	0.019	0.209
XIV	23.62	90.57	60	0.73	5.58	0.019	0.276*
7 May			In bath of liquid helium.				
VI	25.68	90.28	60	1.04	3.16	0.018	0.225
VIII	25.66	90.53	30	0.60	5.24	0.019	0.192
IX	25.62	90.60	30	0.47	5.78	0.019	0.250*
XII	25.86	90.20	60	0.976	2.24	0.018	0.244*
XIII	25.88	90.31	30	0.292	3.28	0.018	[0.422*]
XIV	25.81	90.40	30	0.663	4.09	0.018	0.175
XV	25.77	90.47	30	0.55	4.75	0.019	0.213*
XVI	25.75	90.54	30	0.52	5.34	0.019	0.226

TABLE III.
Atomic Heat Capacity.

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[1]) The difference between c_p and c_v is negligible.

KEESOM and KAMERLINGH ONNES in the liquid hydrogen range. The agreement is relatively satisfactory, considering our calculated accuracy.

The broken line denotes the values for the atomic heat capacity (at constant volume) as calculated from the formula of DEBIJE with $\theta = 88$.

In the first place we note that with lead there is no indication that at the transition to the super-conductive state there occurs a discontinuous change in the atomic heat, even as KAMERLINGH ONNES and HOLST found with mercury.

Further it appears from our measurements that the atomic heat of lead at the lower temperatures is much greater than that calculated from the formula of DEBIJE, employing a value of θ which reproduces approximately the observed values at higher temperatures.

The first indication of this deviation in the case of lead was observed in the results of the measurements of KEESOM and KAMERLINGH ONNES (l.c.) which at the lowest temperatures obtainable with liquid hydrogen indicated that the atomic heat of lead is greater than would be calculated from the formula of DEBIJE with $\theta = 88$ (see their Fig. 10, as well as Fig. 2 of this communication); or otherwise expressed, the value of θ below $18°$ K. gets somewhat smaller than 88 (see Fig. 11, Communication No. 143). Also SIMON l.c. at the temperatures 9.85 to $13.37°$ K., finds θ smaller than 88.

At the temperature of liquid helium this deviation has become so great that the atomic heat there is several times greater than that calculated from the formula of DEBIJE with $\theta = 88$.

A similar behaviour in the case of mercury, since confirmed at temperatures above $9°$ K. by SIMON [1]), was found by KAMERLINGH ONNES and HOLST (l.c.), in connection with the measurements of POLLITZER [2]). A further investigation will be immediately undertaken to see whether this behaviour is also characteristic of substances which are not super-conductive, for example, bismuth.

It is not clear from our measurements whether one would expect that, in approaching $0°$ K., the atomic heat capacity approaches zero or a positive value. It still seems likely from our measurements that the atomic heat capacity approaches zero but at a considerably smaller rate than the T^3 rule of DEBIJE would lead us to expect.

Our results do not show clearly whether the case of lead is explained by the assumption of SIMON [3]) that the deviation, which occurs in the case of gray tin and silicon, is due in these atoms to the existence of two different quantum states with small energy difference. Should this be the case then it would follow from our measurements that $\theta_u = 2.25$ approximately. This would give at $1°$ K. $\triangle C = 0.87$, at $2°$ K. 0.55, at $3°$ K. 0.37. Now it should be observed that several of the more uncertain

[1]) F. SIMON, Zs. f. physik. Chem. **107**, 279, 1923.

[2]) F. POLLITZER, Zs. f. Elektrochemie **17**, 5, 1911; see also **19**, 513, 1913.

[3]) F. SIMON, Berlin Sitz. Ber. 1926, p. 477.

measurements at about 3° K. give rather large values for the heat capacity, 0.7 and 0.4 (see also the values for the core, fig. 1). On the other hand, the more certain measurements give values of about 0.2. It is desirable to secure new measurements in order to be sure of this point.

It is a pleasure to record our hearty thanks to Mr. CHESTER W. CLARK of the University of California for his valuable help in securing the measurements and in calculating the results.

Microbiology. — *On the disappearance of Bacillus typhi from Water.* By Mrs. N. L. WIBAUT—ISEBREE MOENS. (Communicated by Prof. C. PH. SLUITER.)

It is a well known fact that when a culture of bacilli typhi is brought into water the bacteria vanish from the water after a certain time. However no definite information is to be found in the literature about this question as several authors give widely different figures for the period of time after which the bacteria have disappeared. Different circumstances may influence the survival of bacteria in water :

a. physico-chemical conditions : the water may be considered as a medium, in which the growth of the Bacillus typhi is inhibited or aided by the amount of dissolved organic matter, the amount of dissolved oxygen and carbonic acid, the salts, the temperature of the water, the sunlight that penetrates into it, etc. ;

b. biological conditions : in the water there may be organisms devouring the bacteria or acting upon them in one way or another. These organisms will in their turn be affected by the physico-chemical conditions mentioned sub *a.* The oxygen, for instance, may act directly upon the typhus bacilli and also upon the protozoa, which devour the bacteria. [1]

If the difference in lifetime of Bacilli typhi are caused by internal conditions such as oxygen, light etc., then it would be expected that in water of different origin the bacteria should behave in the same way other circumstances being the same. On the other side if the bacteria do not show the same behaviour in different samples of water, the cause of the vanishing of the bacteria is not to be found in external circumstances but in the special properties of the water itself.

During our investigation on water purification we had occasion to study the importance of certain protozoa for the purification process. We got the impression that previous investigators had not recognized sufficiently the rôle of protozoa in the phenomenon of the vanishing of typhus bacteria. We therefore started a series of experiments where in water samples of different origin were inoculated with an equal amount of bacteria other circumstances being alike. At regular intervals the amount

[1] VAN LOGHEM en RUYS, Tijdschrift voor Geneeskunde, 1926, November, Septic tank en typhophage protozoën.

of bacilli typhi was established at the same time te number and the species
of the protozoa were determined by FRANÇOISE VERSCHAFFELT, assisting me
for this biological work.

On 8 February four samples of water of different origin were taken :

a. from the swimming pool Heiligeweg, Amsterdam ;
b. from the Vecht-watersupply ;
c. from a ditch in the province Noord-Holland ;
d. from a cistern, indicated as rainwater.

These four samples of water had been equally infected by a suspension
of typhoid bacilli, recently isolated from faeces. In each flask of a capacity
ot 2 litres and filled with water we put so much of the suspension that the
liquid was faintly opaque. We had previously determined the titre of
protozoa in each flask.

Every three days after the inoculation we determined the number and
species of protozoa; at the same times the presence of bacilli typhi was tested
by Dr. H. PLANTEN in this way that 10 cc. of the water was centrifugated
during 10 minutes, the precipitate being sown on an endoplate. When on
a certain date no bacteria could be found the testing was repeated after
another three days and only if this second test was negative we concluded
that the bacteria had really vanished.

The subjoined tables shows the content of protozoa of each of the water-
samples and the species being present in the water before the inoculation.
In rainwater protozoa could be made out in 0.01 cc. before the
commencement of the experiment ; in ditchwater and the water from
the swimming pool (Heiligeweg) only in 1 cc. while none were found in
1 cc. of the watersupply Vecht, in 10 and 40 cc. however, they were present.

On the 8th of February the flasks were inoculated, kept on room tempe-
rature and in daylight. On the 12th the cultures were inspected for the
first time; in each sample typhus bacilli were present but the number of some
species of protozoa had allready increased while other ones had disappeared.
In the swimming water and in the Vechtwater they have augmented, likewise
in the ditchwater, but in the rainwater the number, as well as the species had
diminished. Table II shows the species of protozoa for each sample of
water ; the following symbols have been used : . for a few, .. for several ;
... for frequent ; .... for many ; :::: for abundant. Tables I and II
present a clear survey of the results of the protozoa-calculation on 8, 12,
15, 18, 22, 25 of February, 1, 5, 8 and 12 of March. Total absence of Bacillus
typhi is designated by ooooooooo.

It must be kept in mind studying the results of these experiments that
mainly the bacteriophage species of protozoa are detected. Of course
besides the bacteriophage protozoa there may occur other plankton forms.
But they dont effect the results of this research. As the typhus bacilli
brought into the water are an excellent food for the bacteriophage protozoa
they will increase abundantly, when there are no more bacteria available
they will decrease.

TABLE I. Febr. 8 — March 12 1926.

	Protozoa present in:	February						March			
		8	12	15	18	22	25	1	5	8	12
Water from Swimming pool Heiligeweg *)	1 cc	+	+	+	+	+	+	+			
	0.1	—	+	+	—	±	—	—			
	0.01	—	+	—	—	—	—	—			
	0.001	—	—	—	—	—	—	—			
Water from the Vechtwater supply	1 cc	—	+	+	+	+	+	+			
	0.1	—	—	+	+	+	—	+			
	0.01	—	—	+	+	—	—	—			
	0.001	—	—	+	+	—	—	—			
Rain-water	1 cc	+	+	+	+	+	+	+			
	0.1	+	+	+	+	+	+	+			
	0.01	+	—	—	+	+	+	—			
	0.001	—	—	—	+	—	—	—			
Ditch-water	1 cc	+	+	+	+	+	+	+	+	+	+
	0.1	—	+	+	+	+	+	—	±	±	—
	0.01	—	—	+	—	—	—	—	—	—	—
	0.001	—	—	—	—	—	—	—	—	—	—

*) Swimming pool replenished February 3.

In water from the *Vechtwatersupply*, although being poor in bacteria as well as in protozoa the bacilli typhi have very soon disappeared : after ten days the water was found to be free from them. Table I shows protozoa being absent in 1 cc. of water at the beginning on 8 February, increasing on 12th and 15th till more than 1000 in 1 cc. Comparing these figures with those on Table II it is clear that Cercobodo bodo [1]) and Alexeieffii and Oicomonas termo have been important. They decrease as soon as Bacilli typhi have disappeared, whereas Pleuromonas jaculans being a non bacteriophage protozoa persist.

Swimming-water behaves towards typhus bacilli still more agressively. After a week the bacteria have already vanished. Here also the Oicomonas termo and Cyclidium glaucoma have increased, Cercobodo Alexeieffii

[1]) For figures see : N. L. WIBAUT, De ontwikkeling van protozoën in hooiinfusie, and also: Protozoën in tankwater van stoomschepen. Tijdschr. v. Microbiologie 1927.

flourishing more afterwards (table II). After bacilli typhi have disappeared, Oicomonas termo can no more be detected, Cercobodo Alexeieffii soon declines. Amoeba limicola, is to be find in the absence of bacillus typhi.

In *ditch-water* the typhus bacilli survive during a longer time than in the

TABLE II. Febr. 8 — March 12 1926.

Species	February							March			
	8	12	15	18	22	25	1	5	8	12	
Swimming water	40cc and 10cc / 1cc										
Amoeba limicola *)	... ...		:::: o ...		...		....				
Actinophrys vesiculata					...						
Bodo minimus	...										
Cercobodo Alexeieffii	....		:::: o ..								
Cinetochilum margaritacea	.										
Collodictyon triciliatum	..										
Cryptochilum nigricans	....										
Cyclidium glaucoma	....	....	.... o		....	::::					
Glaucoma scintillans	.										
Halteria grandinella	.										
Holophrya simplex	.										
Loxophyllum lamella	.										
Monosiga ovata	.										
Oicomonas termo	.... ::::	::::									
Sphaerophrya magna	.										
Vechtwater supply											
Amoeba limicola	.			o							
Bodo minimus	.			o							
Cercobodo Alexeieffii			....	.... o							
" bodo	....	::::		o ...							
Cyclidium glaucoma	....			o							
Oicomonas termo			::::	:::: o	...						
Pleuromonas jaculans			..	o ::::	....	::::					

*) The species occurring in the cultures slightly differs from the description in the literature.

TABLE II.   Febr. 8 — March 12 1926 (*Continued*).

Species	February						March			
	8	12	15	18	22	25	1	5	8	12
Rain-water	40 cc and 10 cc	1 cc								
Amoeba limicola	....			·	°°°°					
Actinophrys vesiculata	...									
Cercobodo Alexeieffii	....	....	·	..	..					
„ bodo		..								
Cyclidium glaucoma	::::									
Loxophyllum lamella	...									
Oicomonas termo		::::	::::	....	::::					
Pleuromonas jaculans		::::		::::	:::: ::::	::::	::::	::::		
Ditch-water										
Actinophrys vesiculata	..	...		..					°°°	
Amoeba limicola	...	..	..	....	...	::::	....	..	.... °°°	....
Astasia Dangeardii	·								°°°	
Aspidisca lynceus	..								°°°	
Bodo minimus	..					·			°°°	
Cercobodo Alexeieffii	...	....	....	....	·				°°°	
Cinetochilum margaritacea	·								°°°	
Chilodon cucullulus	·		·						°°°	
Cryptochilum nigricans	..								°°°	
Cyclidium glaucoma	....			....	·	::::	..	....	.... °°°	.... ..
Didinium balbiani	·	·				·			°°°	
Monosiga ovata	·				·				°°°	
Oicomonas termo		....	....						°°°	
Paramaecium aurelia	...		..	....					°°°	
Pleuromonas jaculans		..	::::	::::		..	....	....	... °°°	
Pleuronema chrysalis	·								°°°	
Rhynchomonas nasuta					..				°°°	
Sphaeroeca volvox	·								°°°	
Vorticella campanulata	..								°°°	

other samples of water. In this experiment they have vanished after 28 days. Among the different protozoa being present in the water at the beginning on 8th of February some bacteriophage species are seen to gain the upper hand. On the 15th of February Cercobodo Alexeieffii and Cyclidium glaucoma are still numerous, as table II shows whereas Oicomonas termo, which appeared so active in the other three samples of water has already disappeared. On the first of March typhus-bacilli can still be made out and Cyclidium glaucoma is present as sole bacteriophage protozoa. It is not quite clear why Bacillus typhi holds on untill 8 March, while the number and species of protozoa remains the same.

On the 2d of March similar experiments were arranged with other samples of water of the same origin in order to see whether the results should confirm those of February.

Tables III and IV illustrate the behaviour of the four samples of water which is nearly the same as that of the previous experiments.

B. typhi disappeared first in *Vechtwater* viz. within six days; again a regeneration of Oicomonas termo is to be observed and a disappearance of this species as soon as the typhus bacilli have gone, as well as a persistence of Pleuromonas jaculans the non-bacteriophage flagellate.

The *swimming-water*, which is by the way originally Vechtwater, after being inoculated, reveals a revival of two species, Cyclidium glaucoma and Cryptochilum nigricans, both good bacteriophages. It strikes us that these species maintain themselves also in the absence of the typhus bacilli. This can be accounted for only by assuming that the water contained a sufficient number of coli-like bacteria from the beginning. These bacilli still present after bacilli typhi have vanished keep, we suppose, the protozoa alive.

Rainwater after being inoculated on the 2d of March, does not contain any typhus bacilli after ten days. Oicomonas termo and Bodo Alexeieffii have augmented and disappear together with the typhus bacilli. Amoeba limicola and Pleuromonas jaculans persist as in the other experiment.

Another experiment was arranged for *ditchwater* viz. a second flask was filled with water which was filtered and had been inspected for the absence of protozoa. The flasks with ditchwater and with filtered ditchwater, the later being absolutety free from protozoa, were now inoculated with typhus bacilli. From the *ditchwater* the bacteria had disappeared after 13 days. Especially Cercobodo Alexeieffii and Cyclidium glaucoma have developed. They persist even after typhus bacilli have vanished. Out of the *filtered ditchwater* however the bacilli typhi have vanished after 17 days. One should expect that if the rôle of the protozoa is really important the bacilli would survive much longer in water without these organisms. As it is evident that in the three other samples of water protozoa are really the deciding factor one must put the question otherwise i.e. why do bacilli typhi vanish so slowly in normal ditchwater in spite of the abundance of protozoa, so that there is a difference of only four days with the behaviour of the bacilli typhi in filtered water where there are no protozoa at all at

30

work ? It seemed reasonable to suppose that the property of the water itself has to do something with this phenomenon.

Supposing this we took water from different ditches in Noord-Holland. One ditch was wellknown to us as there live along people under which febris typhoidea is endemic, on the other ditch this was not so. Two samples from this origin were treated just the same as above. I tabulate the results although they have not as once solved the propounded problem. It appears from table V that the water from these ditches is much more

TABLE III. March 2 — 24 1926.

Water	Protozoa present in:	March					
		2	8	12	15	19	24
Swimming water *)	1 cc	+	+	+	+	+	
	0.1	−	−	+	−	+	
	0.01	−	−	−	−	−	
	0.001	−	−	−	−	−	
Vechtwater supply	1 cc	+	+	+	+		
	0.1	−	+	+	+		
	0.01	−	+	+	+		
	0.001	−	−	−	~		
Rain-water	1 cc	+	+	+	+	+	
	0.1	+	+	+	+	+	
	0.01	+	+	+	−	+	
	0.001	−	+	−	−	−	
Ditch-water	1 cc	+	+	+	+	+	+
	0.1	±	+	+	+	+	+
	0.01	−	−	+	−	+	−
	0.001	−	−	−	−	~	−
Ditch-water (filtered)	1 cc	~	−	~	~	−	−
	0.1	−	−	−	−	−	−
	0.01	−	−	−	−	−	−
	0.001	−	−	−	~	−	~

*) Swimming-pool had been refilled on February 17.

TABLE IV. March 2 — 24 1926.

Species	March						Water
	2	8	12	15	19	24	
Cyclidium glaucoma			::::	....	::::		Swimmingwater (Heiligeweg)
Cryptochilum nigricans		::::	....	....	....		
Amoeba limax		..		..			
Cercobodo Alexeieffii			..				Vechtwater
Oicomonas termo		::::					
Pleuromonas jaculans		....	::::	::::			
Amoeba limicola	not examined		....				
Bodo Alexeieffii		...					Rain-water
Oicomonas termo		::::	....		....		
Pleuromonas jaculans		..	::::	::::	::::		
Amoeba limicola			::::	..	....	...	
Cercobodo Alexeieffii		::::					
Cercobodo Bodo						...	
Cyclidium glaucoma		....	::::	::::	::::	....	Ditch-water
Paramaecium aurelia		...					
Rhaphidiophrys intermedia			..				
Urostyla Weissei			....				

agressive than that of the experiments on 8th February and 2d March. Ditchwater from *Watergang*, where febris typhoidea is endemic, destroys the inoculated typhus bacilli in 5 days, that of Ransdorp after 9 days.

The water from Watergang contains five typhus-eaters indicated on table VI by the letter *T* behind the name. There are three in the water from Ransdorp. In both samples they suffice to destroy the typhus-bacteria, anyhow they are more agressive than in the experiment of 2 March.

The different behaviour of the ditchwater in February and March and in May cannot be explained without further experiments.

One fact is clear from the tables of the grouping of the species of protozoa in these experiments (table II, IV, VI) viz. that in ditchwater approximately the same number and the same species of bacteriophage protozoa survive, even after the disappearance of the typhus bacilli. In rainwater and in Vechtwater not only their number soon decreases but most often they

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TABLE V. May 6—29 1926.

Protozoa present in :	May								
	6	8	10	11	12	15	19	22	29
Ransdorp									
1 cc	+		+	+	+	+	+	+	+
0.1	+		+	+	+	+	+	+	±
0.01	−	typhus inoculation	−	+	+	+	+	±	−
0.001	−		−	−	—	+	−	−	−
Watergang									
1 cc	+		+	+	+	+	+	+	+
0.1	+		+	+	+	+	+	±	±
0.01	+		+	+	+	+	+	−	−
0.001	−		±	−	±	+	−	−	−

disappear absolutely in absence of bacilli typhi. It is evident that here the inoculated bacteria are a welcome and superfluent feeding for the bacteriophage species, always present, though not frequent at the beginning in this water, being very poor on other bacteria. In swimming-water which has been infected with a great number of bacteria by the swimmers, we noticed also a survival of bacteriophage protozoa after the disappearance of typhus bacilli. In a water originally poor in bacteria the inoculated bacteria will act more stimulating on the hungry bacteriophage individuals, than in ditchwater or swimmingwater in which other bacteria are abundant. Here the inoculated bacteria make no such an effect on the bacteriophage species as they had already more than sufficient other bacteria to feed on.

In the water from Ransdorp and Watergang we therefore determined the total number of bacteria being present before inoculating with typhus bacilli. It is to be noticed in the following state that their number is really

Number of bacteria in 1 cc. of ditchwater.

	Culivated on agar at 37° C.	Culivated on agar at 22° C.	Culivated on gelatine at 22° C.
Watergang	7.400	22.100	790.000
Ransdorp	41.600	15.400	innumerable

very high and may be only slightly increased by the inoculated bacilli typhi. On the other hand if bacillus typhi has vanished from the ditch-water the protoza find still a great number of other bacteria to feed on. It is not probable that the protozoa in ditchwater, overcharged as it is with all kinds of bacteria, may be able to select the bacilli typhi.

TABLE VI. May 6 — June 3 1926.
Ransdorp.

Species	May												June	
---	6	8	10	11	12	14	15	17	19	22	25	29	3	
Amoeba limax	.		*	*	*	....	*	::::	...	..	..	..		
Cercobodo Alexeieffii	::::					::::		....	....	...	..	...		T
Coleps hirtus						.								
Cyclidium glaucoma	....					::::		::::	....	....	....		....	T
Oicomonas termo	::::					::::		::::	::::	....	..	..		T
Pleuromonas jaculans						.			::::	::::	::::	::::	::::	
Rhynchomonas nasuta						::::								

(Column 8: inoculation with typhus bacilli)

Watergang.

Species	May												June	
---	6	8	10	11	12	14	15	17	19	22	25	29	3	
Amoeba limax	...		*	....	*	::::	::::	*	::::		....	....		
Anisonema variabile	::::			::::		::::	::::		....		....			T
Cercobodo Alexeieffii	::::			::::		::::	::::		::::		....	..		T
Chilodon uncinatus							.							
Coleps hirtus						.	.							
Collodictyon triciliatum	..					.	.						....	
Cyclidium glaucoma	....			....		::::	::::		::::		....	....	....	T
Halteria grandinella				...										T
Loxophyllum lamella						.	.							
Paramaecium aurelia				....										T
Pleuromonas jaculans						....	....		::::		::::	::::		

(Column 8: inoculation with typhus bacilli)

* not tested.

CONCLUSION.

1[0]. Typhus bacilli do not disappear with equal rapidity in water from different origin brought under the same condition.

2[0]. The disappearance of typhus bacilli is attended with a multiplication of definite species of protozoa (Oicomonas termo, Cyclidium glaucoma and Cercobodo Alexeieffii).

3^0. The rate of disappearance appears to be less dependent on the total number of protozoa present at the beginning of the experiment than on the presence of certain bacteriophage species.

4^0. Ditchwater from Noord-Holland behaves less agressively towards inoculated typhus bacilli than swimming-, rain-, or pipewater.

We purpose to ascertain whether the cause of this aberrant behaviour of ditchwater is to be looked for in its physico-chemical or in its biological properties.

<div align="right">

From the Laboratory of the Municipal Service of
Public Health at Amsterdam.

</div>

Amsterdam, Dec. 1926.

Physiology. — *An Anatomo-physiological study of the Supra-vestibular Tractus in Columba domestica*. By L. J. J. MUSKENS. (Communicated by Prof. J. W. VAN WIJHE).

§ 1. *The function of the tr. strio-mesencephalicus and of the nuc. spiriformis.*

In pursuing the line of research of LONGET, SCHIFF, and others we established in a previous publication [1]), that the commissura posterior in mammals (cat, dog, rabbit) marks the point, at which the physiological effect of the hemisection of the brain-stem is reversed. This may be observed best when, performing in a rabbit a series of hemisections of the brain-stem, starting in the vestibular region, and terminating in the thalamus; then it will be seen that e.g. after severance of the right side of the brain-stem the animal performs a circus-movement towards the left, while the rolling-movement (and also the so-called Magendie Hertwig strabismus, and the tendency to fall down) takes place towards the impaired side. With hemisection, oral to the posterior commissura, the circus-movement takes place towards the unimpaired side. By comparative experiments with subsequent osmic acid staining of the preparations it could be determined in quadrupeds, that only the middle-, and the lateral part of the posterior longitudinal bundle represents the anatomical substrate of these forced movements. However, with lesion oral to the commissura posterior these forced movements and positions occur only if the tertiary vestibular tracts, demonstrated by the present writer. (which connect the supravestibular-commissural nuclei with the globus pallidus and other palaeostriatal nuclei) have been injured. MARCHI's staining presents great difficulties for a precise decision as to which of the superposed nuclei from the region of the commissura-posterior, are the carriers of these functions. These nuclei had been examined purely anatomically by CAJAL, KÖLLIKER, KAPPERS, MESDAG, BECCARI, CASTALDI. In virtue of a number of lesions in the commissural-region in the cat, the present writer established with such a lesion the regular occurrence of bundles descending into the longitudinal bundles, viz. 1^0. of the tr. commissuro-medullaris and 2^0. of the tr. interstitio-spinalis.

A comparative study of the consequences of a number of lesions induced us to correlate the lesion of the first tract with the circus-movement (to

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[1]) Verhandelingen der Koninkl. Akad. van Wetensch. 1902, Deel 8, N⁰. 5; BRAIN, 1914 and 1922.

the impaired side), that of the second with the rolling-movement (towards the unimpaired side).

These data, which were in part new, postulating *inter alia* a vestibular function of the globus pallidus, necessitated a more extensive study of the commissuro-pallidal tracts, as well in their anatomical as in their physiologico-anatomical aspects. As to the former RIESE issued from the Senckenberg Institute at Frankfort an inquiry into the relations in aquatic mammals. The connection of the globus pallidus with the nuclei in the region of the posterior commissure, which had been demonstrated by dint of pains-taking physiological experiments in the rabbit, the dog and the cat, was found very distinctly in these animals in the bundle H. 2. As RIESE has it, RIESE's finding in the aquatic mammals supports "vom vergleichend-anatomischen Gesichtspunkt, die neueren Ergebnisse, zu den MUSKENS auf Grund experimenteller Untersuchungen gelangt ist" [1].

On this ground pursuance of the same anatomo-physiological research into the bird's brain promised a priori to be successful, because in this group of animals with their double mode of locomotion the system of superposed vestibular nuclei in the neighbourhood of the commissura posterior must be extremely developed. I was all the more inclined to do this, because the very accurate investigations by WALLENBERG, and MESDAG's painstaking degeneration work, served as a guide for my experimentation.

While passing by in silence lesions in various parts of the corpus striatum, which did not reveal forced movements, I intend to report my findings with a pigeon that displayed until death circus-movements towards the impaired side. In this animal I had injured the occipito-parietal portion of the forebrain [2] (Fig. 1 and 2 top on the right hand) the portion, that, according to FERRIER, BOYCE and WARRINGTON [3]) and KALISCHER [4]), MAC KENDRICK, JASTROWITZ, GALLERANI and LUSSANNA, elicits on an electric stimulus an eye-movement towards the opposite side. It was especially the tr. strio-mesencephalicus that was found thickly degenerated, (Fig. 1—4) [5]).

The degenerated bundle helps to constitute the ventral part of the brain-stem, bends in the oral part of the thalamus in dorso-lateral direction round the tractus thalamo-tectalis and the round nucleus. It is made up of rather thick fibres, and soonest develops medullary sheaths (SCHROEDER).

Already on this level degeneration is more extensive in the lateral (macrocellular) portion of the nuc. spiriformis, into which nucleus the

[1]) Zeitschr. f. d. Ges. Neur. u. Psych. **90**, 1924, p. 597.

[2]) The small lesion at the bottom fig. 1 marks the most caudal point of the lesion.

[3]) Philos. Transactions Royal Society London B, 1899, Vol. **191**, p. 308.

[4]) Abhandl. der Akad. der Wissenschaften, Berlin 1905, p. 69.]

[5]) This case instances a whole set of experiments. Similar results were achieved in other pigeons, in which a puncture in the forebrain had pierced only or principally the tr. strio-mesencephalicus.

degenerated bundle merges there where the round nucleus begins to disappear, at the oral side of the commissura posterior. In the same areal weak products of decomposition may be found distad, as far as the level

Tr. strio-mesenc.
+metathal.
ventr.

Laesio

Tr. septo-mesenc.

Fig. 1.

Tr. strio-thal.dors.

Tr. strio-mesenc.
+metathal.

Laesio

Tr. occipito-mesenc.

Chiasma

Fig. 2.

Commiss.intratectalis

Commiss. posterior

Nuc.praetectalis

Nuc. spiriformis.
(parb lateralis)

Tr.strio mesenc.
+metathalamicus
ventralis.

Tr. tecto-thalamicus

Tr. quinto-frontalis

Decussatio supra-optica.
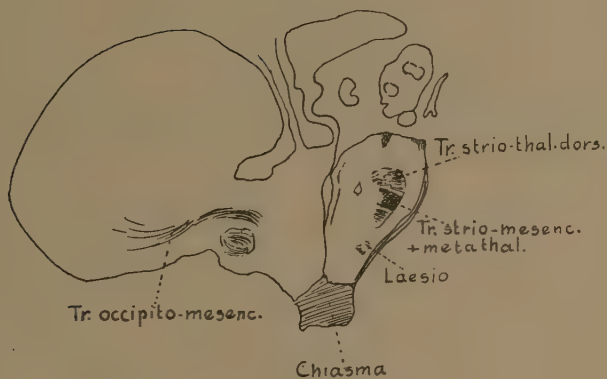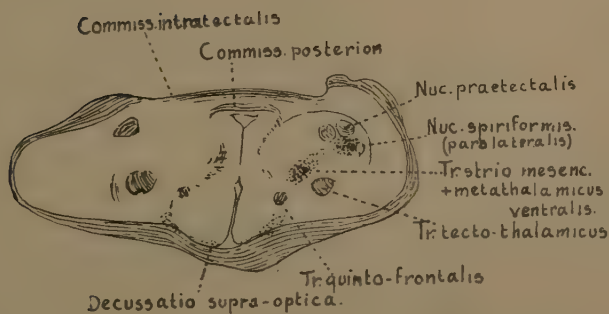
Fig. 3.

of the sensible Trigeminus nucleus, while some of them seem to disappear into the tectum. This descending degeneration (tr. strio-reticularis) was described also by WALLENBERG.
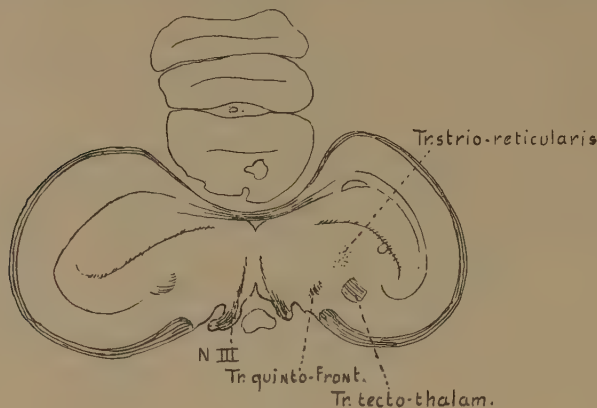

Fig. 4.

In a pigeon (273) the right nuc. spiriformis itself was punctured by a needle. For four days this animal showed a tendency to turn to the right. In the posterior longitudinal bundle no descending degeneration-fibres were encountered.

If this conjecture is correct (partial identity of the nuc. spiriformis of birds with the nuc. commissurae posterior of mammals), this case would warrant the conclusion, that the origin of the fibres in the longitudinal bundle, descending down to the nuc. abducens, is not to be found in the lateral portion of the nucleus.

Neither in my MARCHI-material, nor in the CAJAL-preparations, placed at my disposition by my colleague BOK, do I possess material to disprove WALLENBERG's hypothesis, that also from his nucl. dorso-ventralis (JELGERSMA's nucleus anterior) longitudinal bundle fibres take their origin. Nor is it altogether impossible, that contrary to the quadrupeds, birds with their double mode of locomotion, should have at their disposition two separate mechanisms for these functions.

It seems to me that these findings are to be interpreted in the way alluded to by WALLENBERG and MESDAG (if I am not mistaken), though they do not express that supposition, viz. that the nuc. spiriformis forms part of the system of commissural nuclei.

In birds these nuclei take up a much larger space than in quadrupeds, which tallies with the observation by EDINGER [1]), and WALLENBERG that the fibres of the posterior longitudinal bundle disappear into the nucleus spiriformis. It may be remembered here that already long ago KAPPERS

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[1]) Anat. Anzeiger **33**, 1908, p. 329. EDINGER thought of a connection with the visual function, which may be deemed reasonable.

suspected, that in birds the eye-movements are regulated in the palaeo-striatum (mesostriatum).

In conclusion I think that it may reasonably be assumed that the tractus mesostriato-spiriformis represents one of the two tertiary vestibular bundles, alluded to above, which have been recognized in quadrupeds, namely in their function, and in their descending and ascending fibres, not yet exactly in their initial-, and terminal nuclei in the globus pallidus and in the commissural region.

According to my personal observations made on the cat the fibres of the nuc. posterior thalami (where according to D'HOLLANDER in the rabbit most cortico-thalamic fibres terminate), are directly connected with the two sources of the tractus, which descend in the posterior longitudinal bundle, (viz. nucleus commissurae posterioris and nuc. interstitialis). If, thus, the nuc. spiriformis in birds should occupy partly the place of the nuc. commissurae posterioris of mammals, the question arises, whether in that case the nuc. posterior thalami of mammals may be a newly acquired property of mammals, as a motor nucleus inserted into the cortical tract, (especially subservient to the voluntary eye-movements).

Just as the pallido-commissural bundle in quadrupeds the tr. strio-mesencephalicus of birds degenerates as well in ascending (but in a smaller degree) as in descending direction, which fact had already been established by EDINGER in 1903 and by GROEBBELS. My preparations do not furnish evidence for a partial crossing of this tractus, neither for a connection with the tectum (GROEBBELS).

The wider extent of the area of the commissural vestibular nuclei seems to render it possible that further experiments with various birds may be of use exactly for this more extended analysis, for further localisation of the superposed vestibular nuclei and further homologizing of the mesostriatum and of the commissural region in birds and quadrupeds.

As for our knowledge of the functions of these nuclei, after the function of one of the most significant nuclei has been identified, a way is opened up along which step by step progress can be expected, also with respect to the "Stell reflexe" or righting-reflexes. For it has become evident that a lesion of the secondary, and also of the tertiary ascending vestibular tractus in mammals, as well as in birds entail symptoms, that are comparatively easy to recognize.

§ 2. *Significance of the tracts descending from the mid-brain into the posterior longitudinal bundle, and their relative position in the areal.*

Forced movement in the horizontal plane (circus-, and clock-hand movement with lateral conjugated deviation of the eyes) and forced movement in the plane normal to the long-axis of the animal (rolling movement and skew-deviation of Magendie-Hertwig, tendency to fall down and to lie on one side) are in all animals commonly found to appear

coincidently after lesion of secondary and tertiary vestibular tractus (just as with insects and octopods [1]) after otolith-excision and lesion of the supra-oesophageal ganglion).

This coincidence consists in the circus-movement towards the one side occurring simultaneously with the rolling movement to the other [2]; this is very evident in frogs and tadpoles, and also in fish. As to the latter this may be ascribed to a mechanic, internal connection, (centrifugal force in connection with the labile equilibrium of these animals), for also steamers, when taking a turn on their path to the one side, may be seen to heel over to the other.

The investigation of warm-blooded animals has taught us, that the ascending vestibular tracts, are so disposed within the posterior longitudinal bundle in the areal of this bundle that the fasciculus vestibulo-mesence-phalicus homolateralis and cruciatus in the middle-third part, the fasc. vestibulo segmentalis med. and lat. in the lateral part runs upwards. The middle third part, and that the most medial zone of the areal, is occupied by the descending tr. commissuro-medullaris and the tr. interstitio-spinalis.

When comparing WALLENBERG's [3] classic description of the composition of the posterior longitudinal bundle in pigeons with the later descriptions of that in the rabbit by VAN GEHUCHTEN [4]), and of the same in the cat by the present writer [5]), one is struck by the great similarity e.g. with respect to the increase and the decrease of the bundle in various regions, by the comparative strength of the crossed and the uncrossed bundles, by the ratio of the number of descending fibres from the acusticus-field, even by the shifting of the bundles in the medulla more ventrad, and by the relative position of the bundles (the longest excentric).

However, there is one considerable difference between quadrupeds and birds, in that in birds no trace has yet been found of the absolutely medial position, in quadrupeds, of the bundles descending from the mid-brain.

In connection with the termination of the tr. strio-mesencephalicus in and near the nuc. spiriformis, which on physiological grounds we consider to perform a similar function to that of the nuc. commissurae posterioris in quadrupeds, and in connection with the most lateral position of that nucleus in the formatio reticularis of the mid-brain in birds, an other light is thrown on this discordance. This is borne out by the fact that, whereas in quadrupeds the nuc. commissurae posterioris and the nucleus interstitialis (the two principal sources of the fibres descending into the longitudinal bundle), re'ative to the smaller extent of this region, are not lying far from the raphe, this is, at least partly, quite different in birds.

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[1]) Archiv. f. (Anat. u.) Physiol, 1904, p. 51,

[2]) Journal of Physiology, **31**, 1904, p. 205.

[3]) Anatomischer Anzeiger, **24**, 1903, p. 142.

[4]) Neuraxe, **4**, 1904, p. 63.

[5]) Cf. ·BRAIN, 1914, fig. 7 and 8, p. 370—373; Verslag der zittingen van de Koninkl. Akademie Amsterdam, 19 April 1913, p. 1474.

Here the nuclei in question are larger, and owing to the excentric position of some primary nuclei the fibres descending from them cannot possibly reach in the higher regions the most medial zone in the longitudinal bundle, and only on a lower level do they appear immediately beside the raphe. Figs. 5—9 have been derived from a pigeon, in which the rather considerably lateral incision severed the origin of a bundle, descending into the posterior longitudinal bundle, 10 days before death. They illustrate the anomalous dorsal position of these descending bundles in the pigeon.

Tr. tecto-bulbaris
superfic.

--.Tr. metathalamo-
spinalis.

Fig. 5.

Tr. tecto bulbaris

...Tr. metathalamo-spin.

Corp. trap.

Fig. 6.

Tr. metathal. spin.

Tr. occipito-
+ strio-mesenceph.

Comm. post.

Laesio

N III

Bind arm

Tr. quinto-front.
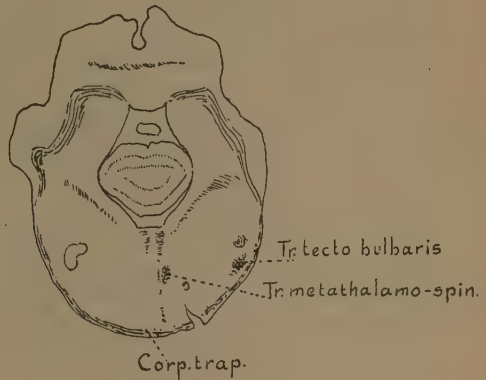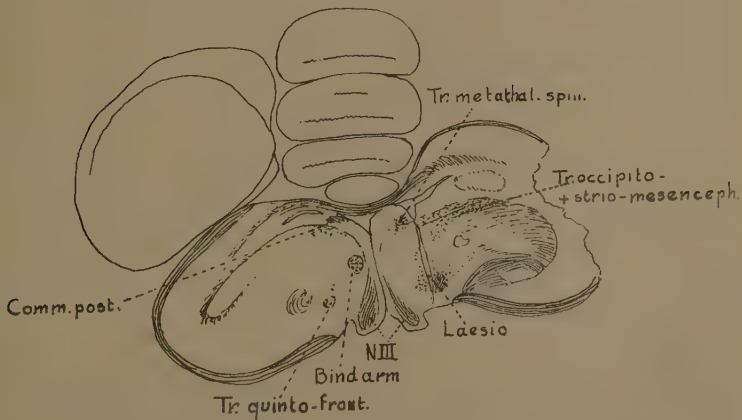
Fig. 7.

From this observation we infer an extremely lateral source of thick longitudinal-bundle fibres. They descend far into the spinal cord just as the tr. interstitio-spinalis, and emanate from a nucleus situated near the

nuc. spiriformis. This, however, cannot take away from the fact, that there exists near the median line a source of similar thick fibres, according to CAJAL.

This is substantiated by BOK's preparations of very young chick-embryos, in which I could verify this latter source of descending bundles-
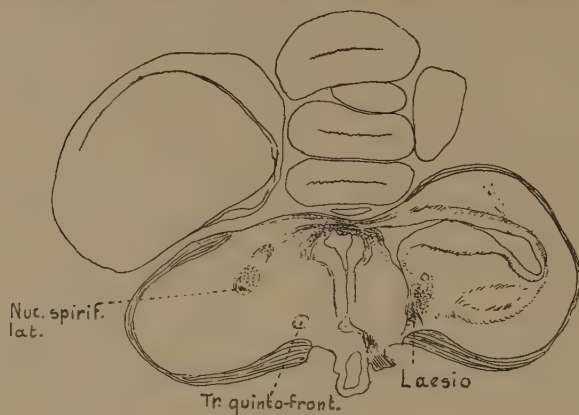
Fig. 8.
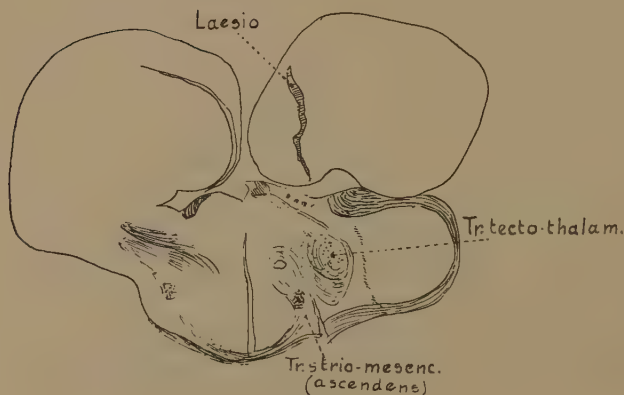
Fig. 9.

called also nuc. interstitialis in birds. This observation also unmistakably bespeaks in birds a double source of descending longitudinal bundle fibres, in connection also with their twofold mode of locomotion.

In connection with the greater extent of these commissural-nuclei in birds there is another physiological point of importance. Theoretically speaking, it should be possible also in warm-blooded animals to make

a lesion in the commissure-region in such a way, that the two forced movements, discussed above, should appear separately, or also in such a way that not the ordinary difference in direction should appear, so that circus-movement to the one side and rolling movement to the *same* side should coincide. Experience, however, teaches, that this does not practically occur in animals, very likely on the one hand because the discussed, ascending vestibular tractus in the longitudinal bundle are lying in the same areal, on the other hand, because the nuc. commissurae posterioris and the nuc. interstitialis are, like their descending bundles, disposed very medially at a short distance from each other. We are justified in assuming that an antero-posterior stab in birds can cut one of the commissural-nuclei, as well as (more caudad) one of the ascending bundles.

This is really the case in the above-named pigeon 22, which received a stab in the commissure from above on the right side. The animal succumbed after 10 days, after it had continually displayed a tendency to move *and* roll towards the left. The sections were stained with osmic-acid, and cut in orderly series.

In consequence of the operation under consideration we find, (figs 7, 6, 5) degenerated at the side operated upon, the bundle (whose course agrees with the tr. interstitio-spinalis (Boyce's bundle), to be much richer in fibres in the pigeon than in the cat. This degeneration proves that, in spite of the extremely lateral lesion, still a nucleus, partly homologous to nuc. interstitialis in the cat, has either been injured through the incision, or is situated more lateral than the lesion, anyhow is severed by the stab into the longitudinal bundle. Quite in keeping with the consequence of such an operation on the right side in cats, the animal invariably rolled towards the left after the dextrolateral operation.

Until the site of origin is exactly determined, this bundle in the pigeon we propose to designate by the name of tr. metathalamo-spinalis.

Whereas after a similar lesion in the cat and in the rabbit circus-movement towards the *impaired* side takes place, as a rule, concomitantly with the rolling towards the *unimpaired* side (and degeneration of the tr. commissuro-medullaris), with this pigeon the circus-movement (and the conjugate deviation of the eyes) had likewise been observed towards the *healthy* side, at the same time no proof was afforded of an injury done to the left tr. commissuro-medullaris. Still, here no doubt that part of the commissura posterior (middle layer) has been severed, in which according to the experiment, as well in birds as in mammals, those secondary ascending vestibular bundles are running of which a lesion always engenders circus-movement towards the uninjured side.

These degenerated commissural bundles can be traced out in the preparations towards the left up to the region of the nuc. spiriformis and the nuc. praetectalis.

From the region of the scar a bundle runs orally to the mesostriatum in the areal of tr. strio-mesencephalicus. In the light of what has been

said in the first paragraph, it is quite natural that we should suppose to have to do here with degeneration of the tr. strio-mesencephalicus, but now of the ascending fibres. These fibres are thinner, less numerous and slightly more ventral in the areal (MUNZER and WIENER) than the descending ones.

As to fish it should be borne in mind, that WALLENBERG [1]) recognized in Selachians termination of the tr. strio-mesencephalicus near the oral end of the fasc. longitudinalis posterior. In cyprinus auratus he traced out a descending bundle from the nuc. lentiformis thalami [2]) down to the caudal part of the medulla oblongata.

About the relations in man of the ascending and the descending vestibular bundles in the longitudinal bundle and in the commissure, little is known thus far. We do know that here also, in contradistinction to the quadruped vertebrates, the bundles descending from the commissural nuclei are not disposed medially. As the assumption is warrantable of the existence, also in man, of a wider development of the superposed, vestibular nuclei, and of the commissural region (erect posture, complicate relations of the eye-movements), it is also reasonable to suppose, that here also considerable extension is answerable for the anomalous position of the descending bundles in the longitudinal bundle.

§ 3. Some of the principal connections of the nuc. spiriformis.

In the pigeon we have identified the nuc. spiriformis as the nucleus, at least as one of the nuclei, that have been inserted into the centrifugal bundle originating in the forebrain and governing the unilateral movement (section of the bundle causes movement towards the side of the lesion). It will now be worth while ascertaining what the anatomical pioneers of the bird's brain have to say about the morphological features of the nucleus, round which our interest has centred in this experiment. Whereas WALLENBERG was struck only by the fact, that the nuc. spiriformis is a terminal stage of longitudinal bundle fibres as well as of a majority of the fibres of the tr. strio-mesencephalicus, it is interesting to observe that MESDAG was obviously struck by the large quantity (6) of the connections of this nucleus found by him, so that he devotes more space to the description of that nucleus than to any of the others, and gives us many particulars concerning them by photographs, and by pictures [3]). The sand-glass-shaped nucleus, then, "consists of medium-sized, triangular or multangular ganglion-cells with exceedingly ramified protoplasm-processess. The cells are packed closely. The axis-cylinders in the centre of the nucleus are not distinguishable from the protoplasm-processes. The nucleus stains very deep with the Cajal method, so that it is very

[1]) Anatomischer Anzeiger, **31**, 1907, p. 395.
[2]) This term is applied to a meta-thalamic nucleus of fishes.
[3]) Loc. cit, p. 107.

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conspicuous in all sorts of series (see photographs 5 and 6). The cells with the deepest stain lie on the anterior margin. The nucleus is joined by all sorts of fibre-tracts to various parts of the mid-brain".

And further on MESDAG says: "There is no doubt but that the Nuc. Spiriformis is extremely significant for the mid-brain. In the paragraph on the optic tract this point will be resumed".

I have devised a diagram exclusively of the tracts, that could be verified by MESDAG after CAJAL's method in chick-embryos, and by WALLEN-BERG after the MARCHI method in pigeons, and by myself after either method.

Undoubtedly the tracts for the vestibular- and the optic impressions are foremost among the reflex-forming connections which govern the direction of the eyes and of the movement to one side, i. e. the movement in the horizontal plane. As regards the former we see for instance rising out of the left vestibular nucleus into the longitudinal

Fig 10.

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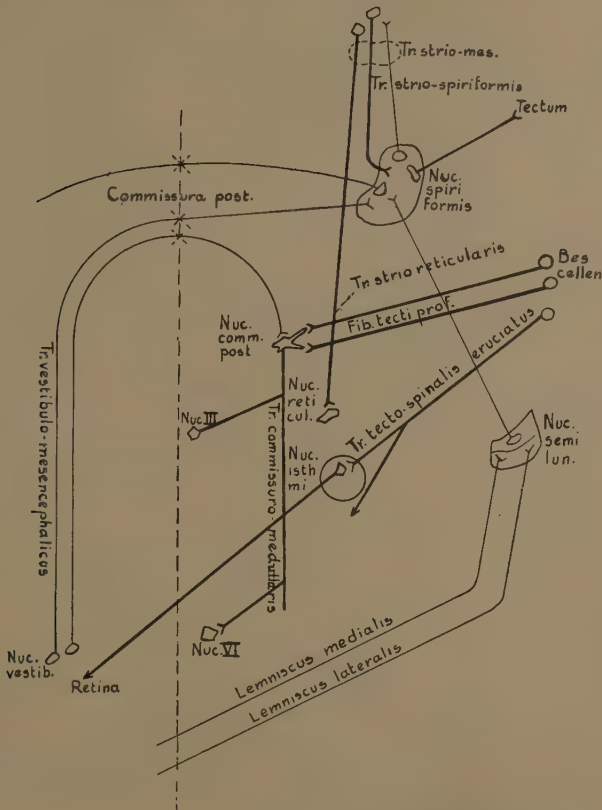
bundle, a tract that (according to CAJAL-preparations of a six-day-old chick embryo) crosses in the commissura posterior, and terminates in the right nuc. spiriformis, and the right nuc. commissurae posterioris. Severance of that bundle (section of the left longitudinal bundle) will render the stimulation of the nuclei concerned insufficient, and will on that account induce circus-movement to the one side, which fits in with the physiological observation (section of the left longitudinal bundle yields circus to the right.

As for the optic stimuli, we know a tractus of the deep tectum fibres, whose axis-cylinders, according to MESDAG are disposed towards the proto-plasm-processes of the ganglion-cells, that constitute the origin of descending fibres in the longitudinal bundle. Where we know, that *lesion* of that nucleus (homologon of the nuc. commissurae post. of mammals) causes the animal to move and look towards the operated side — so mutatis mutandis a *stimulation* of that nucleus : moving and looking towards the other side, thus towards the illuminated field of vision — so here also the anatomical connections tally with the physiological observation, for blinding of one eye yields for a short period a deviation towards the seeing side. We should not omit mentioning here the ganglion Isthmi-retina connection (reflex cone-movements of VAN GELDEREN STORT [1]) according to the amount of illumination. The ventral part of the ganglion isthmi is *pierced* by the tr. tectospinalis non cruciatus, whereas, according to WALLENBERG the Isthmoretinal bundle ends in the crossed retina. So, no contrast here either.

As to the acoustic tractus, CAJAL and MESDAG see the lemniscus medialis terminate at the caudal wall of the tectum opticum, imparting fibres to the nuc. semilunaris medialis. The lemnicus lateralis also enters the nuc. semulinaris medialis, so that given the known tractus of the lobus opticus with the commissural nuclei, the acustical stimuli and tactile stimuli to one side of the body will induce the head and the eyes to move by reflex towards the side of the stimulation.

Anyhow, so far as we know, there is for the moving of head and eyes in the horizontal plane — the only one considered here — no contradiction between the anatomical and physiological data that have thus far been published.

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[1] Archiv. Néerlandaises T., **21**, 1883.

Mathematics. — *Comparison of the Simplified Theories of the Irrational Number.* By Prof. FRED. SCHUH. (Communicated by Prof. D. J. KORTEWEG).

1. Introduction. In an earlier paper (Fusion of the existing Theories of the Irrational Number into a New Theory) I have shown that the different theories of the irrational number may be simplified so that they only consist of the proofs of the following properties:

a. For any two real numbers α and β one and only one of the three relations $\alpha = \beta$, $\alpha > \beta$, $\beta > \alpha$ holds good.

b. If $\alpha > \beta$ and $\beta > \gamma$, we have $\alpha > \gamma$.

c. If α is a real number, there always exists a rational number $> \alpha$.

d. If α is a real number, there always exists a rational number $< \alpha$.

e. If $\alpha > \beta$, there always exists a rational number c, so that $\alpha > c > \beta$.

f. For the system of the real numbers the theorem of the upper boundary holds good.

In this way the further discussion of addition and multiplication is made independent of any special theory of the irrational number and is based on the above mentioned properties.

The aim of this paper is to show how the different theories of the irrational number are connected and to prove the completeness of the system of the real numbers. We shall also give a simple proof of the possibility to extend the system of the rational numbers in such a way that the properties *a.—f.* hold good (cf. n^0. 2—5); this proof, slightly altered, may serve at the same time to demonstrate the completeness of the system of the real numbers (cf. n^0. 13).

2. Building of a system of real numbers. In several ways the system of the rational numbers can be extended to a system for which the properties *a.—f.* mentioned in n^0. 1 hold good. We shall do it here in a way which differs from the usual theories of the irrational number and which has some resemblance to the theories of DEDEKIND and BAUDET.

By a **class** we understand *a set of rational numbers that is not empty, does not contain every rational number, has no greatest number and has the property that a rational number which is less than a number of the set, likewise belongs to the set.* From this it follows immediately *that a number of a class is less than a rational number that does not belong to the class* and *that a rational number which is greater than a*

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rational number that does not belong to the class, does not belong to the class either.

If one of the rational numbers that do not belong to the class is the smallest and if *a* is this smallest number, the class consists of the rational numbers $< a$. In this case we call the class **rational.**

We consider the classes as numbers (*real numbers*) and different classes as different numbers. By assuming *that a rational class must be identified with the smallest rational number that does not belong to it*, also the rational numbers are contained in the new system of numbers.

3. We call a class K **greater** than a class L, *if K contains a number a that does not belong to L.* In this case a number *b* of L is $<$ a, hence a number of K, so that the relation $L > K$ is not satisfied. *Accordingly for two classes K and L one and only one of the three relations $K = L$* (which means that the classes K and L are identical), $K > L$, $K < L$ *holds good.*

If the classes K and L are both rational, with *k*, resp. *l*, as smallest rational numbers that do not belong to them, and if $k > l$, *l* is a number of K that does not belong to L. Consequently $K > L$, *so that the new definition of greater applied to two rational numbers, leads to the same result as the old definition.*

4. *A class K is built of the rational numbers $< K$.*

For if *a* is a number of K, $a < K$, as *a* does not belong to the class formed by the rational numbers $< a$. If, inversely, *a* is a rational number $< K$, there exists a rational number *b* which belongs to K but not to the class of the rational numbers $< a$; accordingly $a \leqq b$, so that *a* belongs to K (because *b* belongs to it).

5. Proofs of the properties mentioned in n^0. 1. *Proof of a.* The validity of *a.* is apparent from n^0. 3.

Proof of b. If K, L and M are classes that satisfy the relations $K > L$ and $L > M$, there exists a rational number *a* which belongs to K but not to L and a rational number *b* which belongs to L but not to M. $b < a$ so that *b* belongs to K. As *b* does not belong to M we have $K > M$.

Proof of c. Let K be a class, *a* a rational number that does not belong to K, and *b* a rational number $> a$. In this case the number *a* belongs to the class of the rational numbers $< b$ but not to K, so that $b > K$.

Proof of d. If K is a class and *c* a number of K, we have $c < K$ (see the property of n^0. 4).

Proof of e. Let K and L be classes that satisfy $K > L$ and let *a* be a number of K that does not belong to L. As K has no greatest number, there is a rational number *c* that is $> a$ and belongs to K. This number *c* does not belong to L. As *a* belongs to the class of the rational numbers

$< c$, we have $c > L$. From the property of n^0. 4 it ensues further that $c < K$.

Proof of f. Let V be a non-empty set of real numbers all of which are smaller than the real number M. We form *the set B of the rational numbers that are smaller than a number of V.* According to *d.* this set is not empty. According to *c.* there is a rational number $> M$; according to *b.* this does not belong to B. If b is a number of B and v a number of V which is $> b$, according to *e.* there is a rational number c so that $b < c < v$; as c is a number of B, B has no greatest number. As moreover a rational number which is smaller than a number of B, evidently also belongs to B, B is a class. We shall now show *that B is the upper boundary of V.*

Let v be a number of V which is $> B$. Then there is a rational number c so that $B < c < v$. This number c belongs to the class B (as $c < v$) so that $B < c$ is in conflict with the property of n^0. 4. *Accordingly a number of V which is $> B$, does not exist.*

Let C be a real number $< B$. There is a rational number a so that $C < a < B$. According to the property of n^0. 4 a is a number of the class B so that a is smaller than a number of V. *This number of V is also greater than C.*

6. Similar systems. *A system of real numbers which contains all rational numbers and for which the properties a.—e. mentioned in n^0. 1 hold good, we shall briefly call a* **system.**

We shall call two systems S and S' **similar** if such a *(1, 1)-correspondence* can be established between the numbers of S and those of S' that the following two properties are valid:

1^0. *the rational numbers correspond to themselves;*

2^0. *if α and β are two numbers of S so that $\alpha > \beta$, and if α' and β' are the corresponding numbers of S', we have $\alpha' > \beta'$.*

We shall call a correspondence that has this property, a **representation** *of one system on the other.*

The similitude of systems is *commutative* and *transitive.* Every system is similar to itself as we can make every number of the system correspond to itself (**identical** *representation*).

7. *If S and S' are two similar systems, in the representation of S on S' the common numbers of the two systems correspond to themselves. We suppose that for two common numbers the relation "greater" is the same in the two systems.*

Let α be a number that belongs to the two systems and α' the number of S' which correspond to α (as number of S). As α and α' both belong to S', we have $\alpha' > \alpha, \alpha' < \alpha$ or $\alpha' = \alpha$. If $\alpha' > \alpha$ there is a rational number b so that $\alpha' > b > \alpha$. From $\alpha < b$ (numbers of S) follows $\alpha' < b$ (corresponding numbers of S') in contradiction to $\alpha' > b$. In the same

way $\alpha' < \alpha$ leads to an absurdity so that $\alpha' = \alpha$ and, accordingly, α corresponds to itself.

Immediate consequences of this theorem are:

the only representation of a system on itself is the identical one;

two systems cannot be represented on each other in more than one way;

a system is not similar to a real part of itself.

8. Owing to the commutativity and the transitivity of similitude we can form *a set of systems any two of which are similar.* We can unite the corresponding numbers of these systems to one conception and consider this as a "number" of which the said corresponding numbers are the representatives.

For according to what was found in n⁰. 7 no two of these corresponding numbers are different numbers of the same system so that a "number" in the new sense does not get two unequal numbers of an already existing system as representatives. As the relation "greater" remains unchanged by the representation, it does not matter from which system we derive the representatives if we have to judge which number is greater.

In the indicated way *similar systems may be united to one system.* By so doing the difference between similar systems disappears entirely as this difference only consists in the names that are given to the numbers or in the way in which they are indicated.

9. Systems for which the theorem of the upper boundary is valid. We prove:

two systems S and S' for which the theorem of the upper boundary is valid, are similar.

Let α be a number of S and A the set of the rational numbers $< \alpha$. In S the number α is the upper boundary of A; for in the first place A has no number $> \alpha$; if γ is a number of S that is $< \alpha$, there is a rational number a so that $\gamma < a < \alpha$; hence a is a number of A that is $> \gamma$. The set A has also an upper boundary in S'; we shall call this α'. We shall make this number α' correspond to α.

Let β be another number of S and *suppose $\alpha < \beta$.* In the same way from β we can derive a number β' of S', which we shall make correspond to β. Now we can choose the rational numbers p and q such that $\alpha < p < q < \beta$. As β' is the upper boundary in S' of the set B of the rational numbers $< \beta$, and as q belongs to B, we have $q \leqq \beta'$. As $p > \alpha$ a rational number $> p$ is also $> \alpha$, hence not a number of A; accordingly A does not contain any number $> p$, so that $p \geqq \alpha'$; for $p < \alpha'$ is contradictory to the fact that α' is the upper boundary of A in S'. From $q \leqq \beta'$, $p \geqq \alpha'$ and $p < q$ we may further conclude *that $\alpha' < \beta'$.* Consequently to unequal numbers of S there correspond unequal numbers of S'.

A number a of A is $\leqq \alpha'$; however, $a = \alpha'$ is excluded as A has no greatest number; a number of A is, therefore, $< \alpha'$. If, inversely, a is a rational number $< \alpha'$, A contains such a number a_1 that $a_1 > a$ (as α' is the upper boundary of A in S'); this number a_1 satisfies the relation $a < a_1 < \alpha$, so that $a < \alpha$ and a is a number of A. The set A is, accordingly, the same as that of the rational numbers $< \alpha'$. Hence to any number α' of S' there corresponds a number α of S, viz. the upper boundary in S of the set of the rational numbers $< \alpha'$.

In the indicated way we get a (1,1)-correspondence between the numbers of S and those of S' for which the property 2^0 of n°. 6 holds good. If α is a rational number, α is the upper boundary in S' of the set of the rational numbers $< \alpha$; hence the correspondence has also the property 1^0 of n°. 6, so that *this correspondence is a representation of S on S'*.

10. *If we extend the system of the rational numbers to a system S for which the properties a.—f. of n°. 1 hold good, and if we omit irrational numbers and add irrational numbers so that a system S' arises for which the said properties likewise hold good, the systems S and S' can be represented on each other* (according to the theorem of n°. 9). According to the theorem of n°. 7 *the omitted numbers correspond to the added ones.* This shows that the transition from S to S' only consists in this *that numbers are omitted and afterwards added again under another name*, for instance that the irrational numbers according to the theory of CANTOR are omitted and the irrational numbers according to the theory of DEDEKIND are added.

11. Completeness of a system of numbers for which the theorem of the upper boundary holds good. We have:

to a system S for which the theorem of the upper boundary holds good, not a single number can be added if we want the properties a.—e. of n°. 1 to remain valid after the addition and also the relation "greater" to remain unchanged for two numbers of S.

Suppose that S may be extended to the system S' (with conservation of the properties a.—e. of n°. 1). Let α' be a number of S' that does not belong to S and A the set of the rational numbers $< \alpha'$. This set has an upper boundary α in S; this number α also belongs to S'. As α' does not belong to S, we have $\alpha < \alpha'$ or $\alpha > \alpha'$.

If $\alpha < \alpha'$ and if p is a rational number so that $\alpha < p < \alpha'$, p is a number of A and hence $p > \alpha$ is contradictory to the fact that α is the upper boundary of A in S. If $\alpha > \alpha'$ and if p is a rational number so that $\alpha > p > \alpha'$, it follows from $p < \alpha$ that there is a number a of A greater than p; from $a > p$ and $p > \alpha'$ follows $a > \alpha'$, in contradiction to the definition of A. Hence in both cases we arrive at an absurdity.

12. The theorem of n⁰. 11 shows *that we cannot omit any number from the system S considered there, if we want the properties a.—f. of n⁰. 1 to remain valid.* For the application of the theorem of n⁰. 11 to the system that arises from S through the omission of numbers would lead to a contradiction.

13. Theorem of completeness. We understand by this:

for a system that is not liable to extension if we want the properties a.—e. of n⁰. 1 to remain valid, the theorem of the upper boundary holds good.

We mean, of course, such an extension *that the relation "greater" for the numbers of the original system remains unchanged.*

We shall extend the system S by forming classes. In deviation from n⁰. 2 we shall now understand by a **class** *a set of numbers of S which is not empty, does not contain every number of S, has no greatest number and has the property that a number of S which is smaller than a number of the set, likewise belongs to the set.*

If among the numbers of S that do not belong to the class there is a smallest a, the class consists of the numbers of S which are $< a$.

We shall consider the classes formed in this way as *numbers of a new system S′* where we identify the class of the numbers of S which are $< a$ (a is a number of S) with a. In this way the numbers of S are also contained in S′.

If K and L are two classes, we give the same definition of $K > L$ as in n⁰. 3. In the same way as there it appears *that the definition of greater which is valid in S′, applied to two numbers of S, leads to the same result as the definition which holds good in S.* Accordingly for the system S′ *the property a. of n⁰. 1* is valid.

The validity of the *property b. of n⁰. 1* for the system S′ is proved as in n⁰. 5.

If K is a class, b a number of S that does not belong to K, and c a rational number $> b$, we have $c > K$ (*property c. of n⁰. 1*). If a is a number of K and d a rational number $< a$, we have $d < K$ (*property d. of n⁰. 1*).

Let K and L be two classes so that $K > L$ and let a be a number of K that does not belong to L. The class K contains a number $b > a$. If c is a rational number so that $b > c > a$, we have $K > c > L$ (*property e. of n⁰. 1*).

Let V be a non-empty set of numbers of S′ all of which are smaller than the number M of S′. We shall now build *the set B of the numbers of S that are smaller than a number of V.* Just as in n⁰. 5 it appears *that B is a class* so that B is a number of S′. If v is a number of V which is $> B$, there is a rational number c (hence a number of S) so that $B < c < v$; according to $c < v$ c is a number of B and from $B < c$ it follows that c is not a number of B; consequently $v > B$ is impossible.

If C is a number of S' which is $< B$ and if a is a rational number so that $C < a < B$, a is a number of B, hence a, and also C, is smaller than a number of V. This shows *that B is the upper boundary of V in S'*, so that for the system S' also the *property f. of n⁰. 1* (theorem of the upper boundary) holds good.

As for the system S' the properties *a.—e.* of n⁰. 1 are valid, S' *is a system as defined in n⁰. 6*. It is further evident from the suppositions of the theorem of completeness, *that the systems S and S' are identical*. As the theorem of the upper boundary holds good for S', this is also valid for S.

14. The theorem of n⁰. 13 together with that of n⁰. 11 (of which it is the converse) shows, *that it amounts to the same if we say of a system that the theorem of the upper boundary holds good for it or that a further extension of the system is impossible*. We can also say *that the condition that is necessary and sufficient for the impossibility of a further extension of the system, is the validity for that system of the theorem of the upper boundary*. It follows further from the theorem of n⁰. 9 *that two systems which are not liable to extension* (with conservation of the properties *a.—e.* of n⁰. 1) *are similar*.

15. A system S can be completed in the way indicated in n⁰. 13 to a system for which the theorem of the upper boundary holds good. From this in connection with the above it is evident that:

a system S is a part of the system of the real numbers and a real part or not according as an extension of S (with conservation of the properties a.—e. of n⁰. 1) is possible or impossible, hence according as the theorem of the upper boundary holds not good for S or holds good.

This means *that S is similar to a part of any system for which the theorem of the upper boundary holds good*.

Mathematics. — *Analytical Introduction of the Trigonometrical Functions, adapted to the Geometrical Introduction*. By Prof. FRED. SCHUH. (Communicated by Prof. D. J. KORTEWEG).

1. Introduction. In the geometrical introduction of the trigoniometrical functions (by the aid of a circle with radius 1) the idea "length of an arc of a circle" is supposed to be defined. This length of an arc can be defined as the limit of the perimeter of a *regular inscribed polygon* $A_0 A_1 A_2 \ldots A_n$ if n increases. We can also indefinitely replace n by its double.

With advantage we can replace $A_0 A_1 A_2 \ldots A_n$ by the *circumscribed polygon* $A_0 B_1 B_2 \ldots B_n A_n$ formed by the tangents at the points A_0, A_1, ..., A_n. In this way we get a simpler formula for y_{2n}, to wit $y_{2n} = \dfrac{4n y_n}{2n + \sqrt{4n^2 + y_n^2}}$, where y_n is the length of $A_0 B_1 \ldots B_n A_n$. By putting $y_{2^n} = x_{n+1}$ this becomes:

$$ x_{n+1} = \frac{2^{n+1} x_n}{2^n + \sqrt{4^n + x_n^2}}. \qquad \ldots \ldots \ldots (1) $$

The length s of the arc $A_0 A_n$ (which is supposed $< \frac{1}{2}\pi$) is $\lim\limits_{n=\infty} x_n$. If we put tg $s = x = x_0$, (1) is also satisfied for $n = 0$. We have, therefore, $\lim\limits_{n=\infty} x_n = $ arc tg x where x_n is defined by (1) and $x_0 = x$.

Inversely we can define arc tg x *as* $\lim\limits_{n=\infty} x_n$, which is a purely analytical way of introducing the cyclometrical and trigonometrical functions. In what follows we shall discuss this method more fully.

2. Arc tg x defined as limit of a sequence. We consider the *real sequence* $\{x_n\}$ *which is defined by* (1) *and* $x_0 = x$ *where* x *is a given real number*. In order to indicate to which value of x the number x_n corresponds, we also write $x_n(x)$ in stead of x_n.

We have $x_n(0) = 0$ and $x_n(-x) = -x_n(x)$, $x_n(x) > 0$ for $x > 0$. If $x > 0$, we have $x_{n+1} < x_n$; accordingly the sequence $\{x_n\}$ decreases monotonely; as in this case $x_n > 0$ for any index n, the sequence is convergent. The latter holds also good for $x \leqq 0$, hence *for any real value of* x. *We call the limit of this sequence* arc tg x; accordingly:

$$ \text{arc tg } x = \lim_{n=\infty} x_n(x). \qquad \ldots \ldots \ldots (2) $$

We have arc tg $0 = 0$ and arc tg $(-x) = -$ arc tg x.

3. The derivative of arc tg x. From (1) ensues:

$$\frac{dx_{n+1}}{dx_n} = \frac{2^{2n+1}}{\sqrt{4^n + x_n^2}\,(2 + \sqrt{4^n + x_n^2})} = \frac{4^{n+1} + x_{n+1}^2}{4\,(4^n + x_n^2)},$$

hence:

$$x_n'(x) = \frac{dx_n}{dx} = \frac{dx_n}{dx_{n-1}}\frac{dx_{n-1}}{dx_{n-2}} \cdots \frac{dx_2}{dx_1}\frac{dx_1}{dx} = \frac{1}{1+x^2} + \frac{x_n^2}{4^n\,(1+x^2)}. \quad (3)$$

consequently :

$$x_n'(x) - \frac{1}{1+x^2} = \frac{x_n^2}{4^n\,(1+x^2)} \leqq \frac{x^2}{4^n\,(1+x^2)} < \frac{1}{4^n},$$

so that $x_n'(x)$ *tends uniformly to the limit* $\dfrac{1}{1+x^2}$ (in the interval formed by all real values of x) *if* n *increases indefinitely. As* according to (3) $x_n'(x)$ *is a continuous function of* x for any index n and any value of x, there follows from (2):

$$\frac{d\,\mathrm{arc\,tg}\,x}{dx} = \lim_{n=\infty} x_n'(x) = \frac{1}{1+x^2} \quad \cdots \cdots \quad (4)$$

Consequently arc tg x *is a differentiable function of* x.

The validity of (4) for $x = 0$ means that $\lim\limits_{x=0} \dfrac{\mathrm{arc\,tg}\,x}{x} = 1$.

4. According to (4) the derivative of arc tg x is always > 0; hence arc tg x *is a monotonely increasing function of* x. From this, in connection with arc tg $0 = 0$, it follows *that* arc tg x *is positive when* x *is positive and negative when* x *is negative.*

5. Formula for arc tg a + arc tg b. We consider the function

$$\varphi(x) = \mathrm{arc\,tg}\,x - \mathrm{arc\,tg}\,\frac{x-b}{1+bx};$$

if $b \neq 0$, the value $-\dfrac{1}{b}$ of x, for which $1 + bx = 0$, must be excluded. According to (4) we find $\varphi'(x) = 0$, so that $\varphi(x)$ is constant in an interval where $1 + bx$ does not assume the value 0. If $ab > -1$, $a \leqq x \leqq b$ or $a \geqq x \geqq b$ is such an interval, so that in this case $\varphi(a) = \varphi(b)$. This gives:

if $ab > -1$ *we have:*

$$\mathrm{arc\,tg}\,a - \mathrm{arc\,tg}\,b = \mathrm{arc\,tg}\,\frac{a-b}{1+ab} \quad \cdots \cdots \quad (5)$$

If we replace b by $-b$ this becomes:
if $ab < 1$ *we have:*

$$\mathrm{arc\,tg}\,a + \mathrm{arc\,tg}\,b = \mathrm{arc\,tg}\,\frac{a+b}{1-ab} \quad \cdots \cdots \quad (6)$$

6. The number 4 arc tg 1 is represented by π. By putting in (5) $b = 1$ we find for $x > -1$:

$$\text{arc tg } x = {}^1/_4 \pi + \text{arc tg } \frac{x-1}{x+1}.$$

In connection with the continuity of arc tg x this leads to:

$$\lim_{x = \infty} \text{arc tg } x = {}^1/_2 \pi, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

whence:

$$\lim_{x = -\infty} \text{arc tg } x = -{}^1/_2 \pi \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

7. If in (6) $a > 0$ and if b increases and tends to $\dfrac{1}{a}$, $\dfrac{a+b}{1-ab}$ increases indefinitely. Hence according to (7):

$$\text{arc tg } a + \text{arc tg } \frac{1}{a} = {}^1/_2 \pi \ (a > 0) \ . \quad . \quad . \quad . \quad . \quad (9)$$

Further:

$$\text{arc tg } a + \text{arc tg } \frac{1}{a} = -{}^1/_2 \pi \ (a < 0) \ . \quad . \quad . \quad . \quad (10)$$

From (9) and (10) there follows:
if $-{}^1/_2 \pi < \text{arc tg } a + \text{arc tg } b < {}^1/_2 \pi$, *we have* $ab < 1$.

In the first place this is the case for $a = 0$. If $a > 0$ it follows from arc tg a + arc tg $b < {}^1/_2 \pi$ in connection with (9), that arc tg $b <$ arc tg $\dfrac{1}{a}$, hence $b < \dfrac{1}{a}$, hence $ab < 1$. If $a < 0$ it follows from arc tg a + arc tg $b >$ $-{}^1/_2 \pi$, in connection with (10), that arc tg $b >$ arc tg $\dfrac{1}{a}$, hence $b > \dfrac{1}{a}$, hence $ab < 1$.

Inversely from $ab < 1$ *we can conclude* $-{}^1/_2 \pi < \text{arc tg } a + \text{arc tg } b < {}^1/_2 \pi$; this ensues immediately from the formula (6) which holds good for $ab < 1$.

8. Definition of tg x. As arc tg x is an increasing function, it follows from (7) and (8) *that* arc tg x *does not assume any value* $\geqq {}^1/_2 \pi$ *and no value* $\leqq -{}^1/_2 \pi$. Arc tg x assumes any value y between $-{}^1/_2 \pi$ and ${}^1/_2 \pi$ for one and only one value of x (as arc tg x is continuous and monotone). *This value of x is called* tg y.

For values of x so that $-{}^1/_2 \pi < x < {}^1/_2 \pi$, tg x *is a continuous function of x, increasing monotonely.* We have tg $0 = 0$, tg $(-x) = -$ tg x, tg ${}^1/_4 \pi = 1$ and:

$$\lim_{x = {}^1/_2 \pi} \text{tg } x = + \infty, \qquad \lim_{x = -{}^1/_2 \pi} \text{tg } x = - \infty \ . \quad . \quad . \quad . \quad (11)$$

We suppose that x while increasing tends to $\frac{1}{2}\pi$ and while decreasing to $-\frac{1}{2}\pi$ as tg x has as yet only been defined for $-\frac{1}{2}\pi < x < \frac{1}{2}\pi$.

From $\lim\limits_{x=0} \dfrac{\text{arc tg } x}{x} = 1$ it follows further that $\lim\limits_{x=0} \dfrac{\text{tg } x}{x} = 1$.

9. Addition-theorem for tg x. By assuming arc tg $a = p$ and arc tg $b = q$ we find from (6):

$$\text{tg } (p + q) = \frac{\text{tg } p + \text{tg } q}{1 - \text{tg } p \, \text{tg } q}, \quad . \quad . \quad . \quad . \quad . \quad (12)$$

where p, q and $p + q$ are supposed to lie between $-\frac{1}{2}\pi$ and $\frac{1}{2}\pi$. If this is the case we have $-\frac{1}{2}\pi < \text{arc tg } a + \text{arc tg } b < \frac{1}{2}\pi$, hence $ab < 1$ (see n⁰. 7), so that the formula (6) holds good, hence also (12). We find, accordingly:

for any set of values of p and q for which p, q and $p + q$ all lie between $-\frac{1}{2}\pi$ and $\frac{1}{2}\pi$ the formula (12) *is valid.*

10. Supplement of the definition of tg x. If q tends to $-\frac{1}{2}\pi$ while decreasing, (12), in connection with the second formula (11), gives:

$$\text{tg } (p - \tfrac{1}{2}\pi) = -\frac{1}{\text{tg } p} \quad . \quad . \quad . \quad . \quad . \quad . \quad (13)$$

on the supposition *that* $0 < p < \frac{1}{2}\pi$.

We shall now extend the definition of tg x to arbitrary real values of x so that (13) *becomes of general validity.* As it ensues from (13) that tg $(p - \pi) = \text{tg } p$, this gives:

if x is a number which is not of the form $(n + \frac{1}{2})\pi$ (n integer), according to definition tg $x = \text{tg } x'$; x' *is here the number between* $-\frac{1}{2}\pi$ *and* $\frac{1}{2}\pi$ *for which* $\dfrac{x - x'}{\pi}$ *is an integer and* tg x *has the meaning indicated in n⁰. 8.*

This definition leaves tg x an odd function of x.

11. According to n⁰. 10 tg x *is periodical with a period* π.

In the interval $-\frac{1}{2}\pi < x < \frac{1}{2}\pi$ we have tg $x = a$, where a is a given number, for only one value x_1 of x. Owing to the periodicity of tg x we have tg $x = a$ for $x = x_1 + n\pi$ (n integer) and for no other value of x. We may also express this thus: *it follows from* tg $x = \text{tg } y$ *that $x - y$ is a multiple of π.*

12. *If none of the numbers $p, q, p + q$ is of the form $(n + \frac{1}{2})\pi$ (n integer), the formula* (12) *holds good.*

In order to prove this we may, owing to the periodicity of tg x, assume *that p and q lie between* $-\frac{1}{2}\pi$ *and* $\frac{1}{2}\pi$. The case that also $p + q$ lies between $-\frac{1}{2}\pi$ and $\frac{1}{2}\pi$ is that of n⁰. 9. We may, therefore,

assume *that $p + q$ lies between* $^1/_2\,\pi$ *and* π (in which case p and q lie between 0 and $^1/_2\,\pi$), as the case $-\pi < p + q < -^1/_2\,\pi$ can be reduced to this because $\mathrm{tg}\,x$ is an odd function. Taking (13) into account we have in this case:

$$\mathrm{tg}\,(p + q) = \mathrm{tg}\,\{(p - ^1/_2\,\pi) + (q - ^1/_2\,\pi)\} = \frac{\mathrm{tg}\,(p - ^1/_2\,\pi) + \mathrm{tg}\,(q - ^1/_2\,\pi)}{1 - \mathrm{tg}\,(p - ^1/_2\,\pi)\,\mathrm{tg}\,(q - ^1/_2\,\pi)} =$$

$$= \frac{-\dfrac{1}{\mathrm{tg}\,p} - \dfrac{1}{\mathrm{tg}\,q}}{1 - \dfrac{1}{\mathrm{tg}\,p}\,\dfrac{1}{\mathrm{tg}\,q}} = \frac{\mathrm{tg}\,p + \mathrm{tg}\,q}{1 - \mathrm{tg}\,p\,\mathrm{tg}\,q}.$$

13. Definitions of sin x and cos x. As definitions we give:

$$\sin x = \frac{2\,\mathrm{tg}\,^1/_2\,x}{1 + \mathrm{tg}^2\,^1/_2\,x}, \qquad \cos x = \frac{1 - \mathrm{tg}^2\,^1/_2\,x}{1 + \mathrm{tg}^2\,^1/_2\,x} \quad . \quad . \quad (14)$$

These definitions fall short if x is of the form $(2n + 1)\,\pi$ (n integer). Leaving the *continuity* of $\sin x$ and $\cos x$ intact, also for $x = (2n + 1)\pi$, we find from (14) by taking the limit:

$$\sin (2n + 1)\,\pi = 0, \qquad \cos (2n + 1)\,\pi = -1.$$

It ensues from (14) that $\sin x$ and $\cos x$ are periodical functions with a period 2π, and further, in connection with (12), *that* $\dfrac{\sin x}{\cos x} = \mathrm{tg}\,x$; $\lim\limits_{x=0} \dfrac{\mathrm{tg}\,x}{x} = 1$ leads further to $\lim\limits_{x=0} \dfrac{\sin x}{x} = 1$.

From (12) and (14) we also find the formulas for $\sin (a + b)$ and $\cos (a + b)$ and in the well known way the other properties of $\sin x$ and $\cos x$ can be derived from them.

14. Application of the trigonometrical functions to angles. We can easily prove:

if the numbers a and b satisfy the relation $a^2 + b^2 = 1$ we can always find one and only one number x so that $\cos x = a$, $\sin x = b$, $-\pi < x \leqq \pi$.

From this we can a posteriori derive the connection between trigonometrical functions and angles (which in the elementary trigonometry is the starting point for the definition of these functions). See for this my book: "Het getalbegrip, in het bijzonder het Onmeetbare getal, met toepassingen op de Algebra, de Differentiaal- en de Integraalrekening," §§ 285—290.

Chemistry. — *Investigations into the Constitution of Artificial Ultra-marines.* II. *On Ultramarine-blue with high Silica-content and on Silver-, Silversodium-, Selenium- and Silverselenium-Ultramarines.* By Prof. Dr. F. M. JAEGER and Mr. F. A. VAN MELLE.

§ 1. In connection with our paper, already published in these Proceed-ings [1]), concerning the constitution of artifical and natural *ultramarines*, it was planned by us to substitute successively a part or all of the mobile constituents of these compounds, e.g. their *sodium*-atoms by *Li, K, Rb, Cs, Zn, Ba, Sr, Ag,* etc., or their *sulphur*-atoms by *selenium,* — with the purpose to elucidate the influence of such substitutions upon the ROENTGEN-spectrograms obtained with these substances. In this way it would perhaps be possible to ascertain, which components among those were a real part of the „rigid, periodical structure" of these compounds and which were merely dispersed in a statistical way within that rigid structure.

To proceed quite systematically in this, we have restricted our studies to one species of *ultramarine* only, chosen as primary material, namely to the *ultramarine-blue*-GUIMET N⁰. 7553, which in a pure, thoroughly washed and unground condition was most kindly given us in a consider-able quantity by the *Usines d'Outremer*-GUIMET in *Fleurieu sur Saône.* Its composition appeared to correspond almost exactly to the formula: $Na_6Al_4Si_6O_{23}S_3$; its colour was a vivid, brilliant blue [2]).

For the sake of completeness, in Table I the results of the analysis of a powder-spectrogram of this preparation are put together. As in all sequent cases, an improved cylindrical camera was used, having a diameter of 88,8 m.m., and allowing the photographical film to be attached to its outer side; the camera is completely inclosed in a copper box. This variation of the older instrument has satisfied us in all respects.

In total 33 interference-images were measured; the grating-constant is here: $a_0 = 9,06$ A.U.

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[1]) These Proceedings, **30,** 249, (1927).

[2]) The ultramarine used here is an ultramarine with high silica- and moderate sulphur-content; its spec. weight at (25° C.) was: 2.35. Analysis furnished: 15.4 % Na; 13.0 % Al; 19.2 % Si; 10.6 % S; 41.8 % O, in which is included 2.78 % clay-residue. For $Na_6Al_4Si_6O_{23}S_3$ is necessary: 15.6% Na; 12.3% Al; 19.3 % Si; 10.9 % S; 41.8 % O. In no case the substance can correspond, as HOFFMANN supposed, to the aluminium-sodium-silicate: $Na_4Al_4Si_6O_{20}$: neither does it correspond to the *Marienberger blue* used by HEUMANN, because this was a product poor in silica.

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TABLE I. Powder-spectrogram of Ultramarine-blue: $Na_6 Al_4 Si_6 O_{23} S_3$ (GUIMET—III, N⁰. 7553). (Fig. 1A).

N⁰. of Image	2l in mm.:	Estim. Intensities:	λ:	$\sum (h^2)$:	$\frac{\theta}{2}$:	$sin^2 \frac{\theta}{2}$ (observed):	$sin^2 \frac{\theta}{2}$ (calculated):	Indices:
1	(21.4)	4	α	2	6°54′	0.0144	0.0144	(110) only once observed
2	37.1	10	α	6	11°58′	0.0430	0.0432	(211)
3	43.6	1	β	10	14°4′	0.0591	0.0590	(310)
4	48.2	7	α	10	15°33′	0.0719	0.0720	(310)
5	53.0	8	α	12	17°6′	0.0865	0.0864	(222)
6	55.4	1⁻	β	16	17°52′	0.0941	0.0944	(400)
7	57.1	2	γ	14	18°25′	0.0999	0.1008	(321)
8	59.0	1⁻	β	18	19°2′	0.1063	0.1062	(330) ; (411)
9	61.5	4	α	16	19°50′	0.1151	0.1152	(400)
10	65.4	5	α	18	21°6′	0.1296	0.1296	(330) ; (411)
11	71.3	1	β	26	23°0′	0.1528	0.1534	(431) ; (510)
12	72.9	1	α	22	23°31′	0.1590	0.1584	(332)
13	79.6	5	α	26	25°41′	0.1877	0.1872	(431) ; (510)
14	82.4	1⁻	β	34	26°35′	0.2004	0.2006	(433) ; (530)
15	86.0	1	α	30	27°44′	0.2165	0.2160 {	(521) as β-line (600)
16	89.2	3	α	32	28°47′	0.2317	0.2304	(440)
17	92.0	2	α	34	29°41′	0.2451	0.2448	(433) ; (530)
18	95.0	2	α	36	30°39′	0.2599	0.2592	(600)
19	97.9	2	α	38	31°35′	0.2742	0.2736	(532) ; (611)
20	106.4	3	α	44	34°20′	0.3180	0.3168	(622)
21	109.1	1⁻	β	56	35°12′	0.3322	0.3304 {	(642) as α-line (631)
22	111.9	2	α	48	36°6′	0.3471	0.3456	(444)
23	114.7	1	α	50	37°0′	0.3621	0.3600	(543) ; (505) ; (710)
24	120.0	2	α	54	38°43′	0.3910	0.3888	(633) ; (721)
25	122.1	1	α	56	39°23′	0.4026	0.4032	(642)
26	124.8	1	α	58	40°16′	0.4178	0.4176	(730)
27	130.2	1	α	62	42°0′	0.4476	0.4464	(732)
28	132.8	1	α	64	42°50′	0.4620	0.4608	(800)
29	135.0	1	α	66	43°33′	0.4747	0.4752	(554) ; (741) ; (811)
30	145.5	1	α	74	46°56′	0.5337	0.5328	(750) ; (743) ; (831)
31	148.4	1	α	76	47°51′	0.5498	0.5472	(662)
32	150.9	1	α	78	48°41′	0.5640	0.5616	(752)
33	153.5	1	α	80	49°31′	0.5785	0.5760	(840)

Radius of the Camera: 44.4 mm. $V = 55000$ Volt.

Exposure: 2 hours. $\lambda_\alpha = 1,540$ A.E.; $\lambda_\beta = 1,388$ A.U.

Quadratic Equation: $sin^2 \frac{\theta}{2} = 0.0072 (h^2 + k^2 + l^2)$ (α-radiation)

$$sin^2 \frac{\theta}{2} = 0.0059 (h^2 + k^2 + l^2) (\beta\text{-radiation})$$

§ **2.** To corroborate our conclusions formerly drawn about the type of the fundamental grating and about the space-groups of these compounds, we wish to give here some additional and very persuasive complementary data concerning the minerals *noseane* and *hauyne*. We happened to prepare two rotation-spectrograms of some small, really simple crystals of both minerals, which spectrograms were splendidly developed, containing a great number of interference-images, which could accurately be measured. Moreover, we succeeded in rotating the crystals in these experiments round a principal axis [100], — an experiment which formerly had been

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TABLE II. Rotation-Spectrograms of *Noseane* and *Hauyne*, if rotated round a principal axis [100].

A. *Noseane.* The following images were observed:	**B.** *Hauyne.* The following images were observed:
Principal spectrum:	Principal spectrum:
λ_α : $(0\,\overline{3}\,\overline{1})$; $(0\,\overline{5}\,\overline{1})$; $(0\overline{4}0)$; (031) ; (033) ; $\qquad\qquad\qquad\qquad\qquad (051)$,	λ_α : $(0\overline{1}3)$; (004) ; (015) ; $(03\overline{3})$; $(05\overline{3})$.
λ_β : $(0\,\overline{5}\,\overline{3})$; (053) ; (033).	λ_β : (004) ; (035) ; $(03\overline{3})$.
1st Accessory spectrum:	1st Accessory spectrum:
λ_α : $(\overline{1}\,\overline{3}\,\overline{2})$; $(\overline{1}\overline{5}0)$; (121) ; (132) ; (150) ; (152)	λ_α : $(1\overline{1}3)$; (105) ; (152) ; (121) ; $(13\overline{2})$; $(14\overline{3})$.
λ_β : (150).	λ_β : (121) ; $(14\overline{3})$.
— 1st Accessory spectrum:	— 1st Accessory spectrum:
λ_α : $(\overline{150})$; $(\overline{132})$; $(\overline{150})$; $(\overline{152})$.	λ_α : $(\overline{1}\overline{1}3)$; $(\overline{1}05)$: $(\overline{1}52)$; $(\overline{1}2\overline{1})$; $(\overline{1}3\overline{2})$; $(\overline{1}4\overline{3})$.
λ_β : $(\overline{1}21)$; $(\overline{150})$.	λ_β : $(\overline{1}4\overline{3})$.
2nd Accessory spectrum:	2nd Accessory spectrum:
λ_α : $(\overline{2}\overline{3}1)$; (233) ; (242).	λ_α : (213) ; (215) ; $(21\overline{1})$; $(22\overline{2})$; $(23\overline{3})$; $(24\overline{2})$.
λ_β : (222).	λ_β : (253) ; (235).
— 2nd Accessory spectrum:	— 2nd Accessory spectrum:
λ_α : $(\overline{2}\overline{3}1)$; $(\overline{2}20)$; $(\overline{2}33)$; $(\overline{2}42)$.	λ_α : $(\overline{2}13)$; $(\overline{2}15)$; $(\overline{2}2\overline{2})$; $(\overline{2}33)$; $(\overline{2}42)$.
λ_β : $(\overline{2}22)$.	λ_β : $(\overline{2}2\overline{2})$; $(\overline{2}53)$.
3rd Accessory spectrum:	3rd Accessory spectrum:
λ_α : $(3\overline{3}\overline{2})$; (310) ; (321) ; (332).	λ_α : (312) ; (303) ; (314) ; (310) ; $(32\overline{1})$; $(33\overline{2})$; $\qquad\qquad\qquad\qquad\qquad (350)$.
λ_β : (350) ; (352).	λ_β : $(35\overline{2})$; (350).
— 3rd Accessory spectrum:	— 3rd Accessory spectrum:
λ_α : $(3\,\overline{4}\,\overline{1})$; $(\overline{3}10)$; $(\overline{3}50)$; $(\overline{3}52)$.	λ_α : $(\overline{3}12)$; $(\overline{3}03)$; $(\overline{3}14)$; $(\overline{3}05)$; $(\overline{3}10)$; $(32\overline{1})$; $\qquad\qquad\qquad\qquad\qquad (332)$; $(\overline{3}50)$.
λ_β : $(\overline{3}\,\overline{3}\,\overline{1})$.	λ_β : $(\overline{3}03)$; $(\overline{3}50)$; $(\overline{3}52)$.
4th Accessory spectrum:	4th Accessory spectrum:
λ_α : $(4\overline{3}\overline{1})$; (431).	λ_α : (422) ; (413) ; $(43\overline{1})$; (420).
λ_β : $(4\overline{4}0)$.	λ_β : (404) ; $(43\overline{1})$.
— 4th Accessory spectrum:	— 4th Accessory spectrum:
λ_α : $(\overline{4}\,\overline{3}\,\overline{1})$; (431) ; $(\overline{4}20)$; (422).	λ_α : $(\overline{4}22)$; $(\overline{4}13)$; $(\overline{4}20)$; $(\overline{4}2\overline{2})$; $(\overline{4}31)$.
λ_β : $(\overline{4}\overline{4}0)$.	λ_β : $(\overline{4}3\overline{1})$; $(\overline{4}13)$; $(\overline{4}04)$.

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impracticable. The rotation-spectrograms obtained were graphically ana-
lysed according to BERNAL's method [1]: thus in the case of *noseane* up
to 50, in that of *hauyne* even up to 72 spots of the film, indices were
attributed in this way. The data obtained, arranged after the successive
hyperbolae, are reproduced in Table II.

N⁰. of Image	Estim. intensities:	Radiation:	$2l$ in mm.:	$\dfrac{\theta}{2}$:	$sin^2 \dfrac{\theta}{2}$ (ob- served):	$sin^2 \dfrac{\theta}{2}$ (cal- culated):	Indices;
1	5	α	37.2	12°0′	0.0432	0.0440	(211)
2	5	β; or α	43.6	14°4′	0.0591	0.0596 {	(310) as α-line (220)
3	9	α	48.6	15°39′	0.0728	0.0733	(310)
4	5	α	53.4	17°14′	0.0878	0.0880	(222)
5	4	α	57.5	18°33′	0.1012	0.1025	(321)
6	2	β	59.2	19°6′	0.1071	0.1073	(330) or (411)
7	9	α	66.1	21°19′	0.1321	0.1319	(330) or (411)
8	1	β; or α	81.1	26°16′	0.1958	0.1907 {	(440) as α-line (431); (510)
9	2	β	87.6	28°16′	0.2243	0.2282	(611) or (532)
10	3	α	90.0	29°3′	0.2358	0.2346	(440)
11	5	α	93.0	30°0′	0.2500	0.2493	(433) or (530)
12	3	α	95.7	30°52½′	0.2633	0.2638	(600)
13	4	α	98.6	31°48½′	0.2778	0.2785	(611) or (532)
14	2	α	101.6	32°46½′	0.2931	0.2932	(620)
15	1	β	107.4	34°39′	0.3233	0.3218	(633) or (721)
16	1	β	112.4	36°16′	0.3499	0.3458 {	(730) as α-line (444)
17	5	α	120.9	39°0′	0.3960	0.3958	(633) or (721)
18	2	α	126.1	40°41′	0.4250	0.4251	(730)

TABLE III. Powder-spectrograms of Silver-Ultramarine N⁰. 3 with 46.5 % Ag, prepared from Ultramarineblue-GUIMET III; 1st Fraction.

Radius of the Camera: 44.4 mm. $V = 55000$ Volt. Time of exposure: 3 hours.

Quadratic Equation (α-radiation): $sin^2 \dfrac{\theta}{2} = 0.00733 \, (h^2 + k^2 + l^2)$.

(β-radiation): $sin^2 \dfrac{\theta}{2} = 0.00596 \, (h^2 + k^2 + l^2)$,

$\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U. $\alpha_0 = 8.99$ A.U.
Body-centred, cubic grating. The colour of the compound is pale yellow.

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[1] J. D. BERNAL, Proceed. R. Soc. London, *A*, **113**, p. 117, (1927).

The conclusions formerly drawn are fully confirmed by these data, as in no case the sum $(h + k + l)$ appears to be an odd number. Undoubtedly a bodily-centred grating is, therefore, present in this structure, with $a_0 =$ about 9,1 A.U.

§ 3. Starting from this *ultramarine-blue*-GUIMET n⁰. 7553, now the corresponding *silver-ultramarine* was first prepared in the way described by HEUMANN [1]). 15 gramms of the *sodium-ultramarine* were constantly heated in sealed glass-tubes during 15 hours at 120° C. with 25 gramms *silvernitrate* and 20 ccm. water. The yellowish mass obtained was suspended in water, the solution of *silvernitrate* filtered-off, the residu washed [2]) thoroughly several times and during some time treated on the waterbath with a solution of a little *iodine* in *potassium-iodide*. After removing this solution by filtering, and having washed the residue, this was at 90° treated several times with a 10 % solution of *potassium-cyanide*. This salt being removed by thoroughly washing, the powder was suspended in several litres of distilled water and repeatedly fractionated by sedimentation. In this way the *silversulphide* which has been formed, having much greater specific weight than the derived compound, can be removed almost completely, if this treatment be continued as long as black particles are observed; the fraction which remains in suspension during the longest time, is the purest and was always used in the final experiments. Analysis of this yellow compound gave (n⁰. 3); 46,5 % Ag; calculated for $Ag_6Al_4Si_6O_{24}S_3$: 46,6 % Ag. [3])

Of this *silver-ultramarine* a powder-spectrogram was obtained, which showed a great number of very sharp interference-images: the results of its analysis are collected in Table III.

§ 4. For the sake of comparison, we have in the same way prepared some other *silver-ultramarines* also, starting from other *ultramarines*. Thus, starting from *ultramarine-pink* GUIMET OVR (with 14,25 % Al; 19,33 % Si; 10,4 % S; spec. wgt.: 2,34 at 25° C.) a greyish looking *silver*-compound with 34,4 % Ag; from *ultramarine-blue* N⁰. 13 of the *Vereinigte Ultramarinfabriken*, a *silver-ultramarine* with 34,2 % Ag; and from an *ultramarine-green* N⁰. X of the same origin, a *silver*-derivative

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[1]) K. HEUMANN, Ann. der Chemie, **199**, 253, (1879); **201**, 262, (1880); **203**, 174, (1881).

[2]) This purification appears to be very troublesome, because the *silver-ultramarine* in contact with water, changes, like finely divided clay, into a colloidal, plastic mass, which after some time is transformed into hard, strongly conglomerating clumps. It may, therefore, be used for the fabrication of pottery, which on strongly heating, however, gets brown under loss of sulphur. Perhaps it would be possible, by heating it in the vapours of *alcali-chlorides*, to stain it superficially with a blue colour. This will be tried out afterwards.

[3]) Later-on we will publish the complete analysis of the *silver*-compound. It must be remarked, that the Ag-content appears to be somewhat variable, probably because of no complete homogeneity of the preparation.

Nº. of Image	2l in mm.:	Intensities	λ:	$\frac{\theta}{2}$:	$sin^2 \frac{\theta}{2}$ (ob-served):	$sin^2 \frac{\theta}{2}$ (cal-culated):	Indices:
TABLE IV. Powderspectrogram of Silver-Ultramarine from Ultramarine-green V. U. F. Nº. X. (Fig. 1B)
1	34.6	2	β	11°10′	0.0374	0.0362	(211)
2	37.5	5	α	12°6′	0.0439	0.0446	(211)
3	43.7	3	β	14°6′	0.0593	0.0604 {	(310) as α-line (220)
4	49.7	8	α	16°2′	0.0763	0.0743	(310)
5	53.1	1	β	17°8′	0.0868	0.0846	(321)
6	54.5	4	α	17°35′	0.0913	0.0892	(222)
7	58.9	3	α	18°58′	0.1056	0.1040	(321)
8	60.3	2	α	19°27′	0.1108	0.1179	(400)
9	64.3	1	β	20°44′	0.1252	0.1208	(420)
10	67.0	8	α	21°37′	0.1358	0.1337	(330) and (411)
11	70.8	3	α	22°50′	0.1505	0.1486	(420)
12	74.4	2	α	24°0′	0.1655	0.1635	(332)
13	77.9	4	α	25°8′	0.1804	0.1783	(422)
14	81.2	4	α	26°12′	0.1950	0.1922	(431) and (510)
15	84.1	2	β	27°8′	0.2080	0.2054	(433) and (530)
16	86.3	1	β	27°50′	0.2180	0.2174	(600)
17	90.8	3	α	29°17′	0.2391	0.2378	(440)
18	93.8	5	α	30°16′	0.2541	0.2526	(433) and (530)
19	97.0	3	α	31°18′	0.2699	0.2675	(600)
20	99.7	4	α	32°10′	0.2835	0.2823	(532) and (611)
21	102.6	2	α	33°6′	0.2983	0.2972	(620)
22	105.6	2	α	34°4′	0.3138	0.3121	(541)
23	108.4	2	α	34°58′	0.3284	0.3269	(622)
24	110.7	1	α	35°43′	0.3408	0.3418	(631)
25	113.7	1	α	36°41′	0.3569	0.3566	(444)
26	115.6	1	α	37°17′	0.3670	0.3715	(543) ; (505) ; (701)
27	121.9	5	α	39°19′	0.4014	0.4012	(633) and (721)
28	127.3	2	α	41°4′	0.4315	0.4309	(730)
29	132.5	1	α	42°44′	0.4605	0.4607	(732)

				TABLE IV (Continued).		
No. of Image:	2l in mm.:	Intensities:	λ:	θ/2 :	$sin^2\frac{\theta}{2}$ (observed):	$sin^2\frac{\theta}{2}$ (calculated):	Indices:
30	137.8	1	α	44°28′	0.4906	0.4904	(554); (741); (811)
31	140.4	1	α	45°17′	0.5049	0.5052	(644) and (820)
32	143.2	1	α	46°12′	0.5210	0.5201	(653)
33	145.5	1	α	46°56′	0.5338	0.5350	(660); (822)
34	148.4	1	α	47°53′	0.5503	0.5499	(750); (743); (831)
35	150.9	1	α	48°41′	0.5641	0.5647	(662)
36	153.7	1	α	49°35′	0.5796	0.5795	(752)
37	155.9	1	α	50°17′	0.5917	0.5944	(840)
38	158.8	1	α	51°14′	0.6080	0.6093	(833)
39	164.4	1	α	53°2′	0.6383	0.6390	(655)
40	169.7	1	α	54°45′	0.6669	0.6687	(851)

Radius of the camera: 44.4 mm.

Wavelength: $\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.381$ A.U.; $a_0 = 8.93$ A.U.

$V = 55000$ Volt. Time of exposure: 3 hours.

Quadr. equation: $sin^2\frac{\theta}{2} = 0.00743\ (h^2 + k^2 + l^2)$. (α-radiation).

$$sin^2\frac{\theta}{2} = 0.00604\ (h^2 + k^2 + l^2). \quad \text{(β-radiation).}$$

with 51,0 % Ag. Notwithstanding the differences in *silver*-content, the powder-spectrograms of all these preparations were *rigorously identical*. In Table IV the results of the analysis of the last mentioned spectrogram are reviewed ($a_0 = 8,93$ A.U.), because this showed the greatest number of sharp lines. The relative intensities of the most important, images are, — as far as so rough an estimation allows us to judge it, — analogous to those of Table III.

From these data the following conclusions may be drawn:

1°. Notwithstanding the different Ag-content of the compounds studied, their ROENTGEN-spectrograms appear to be practically identical.

2°. The original periodical structure of the *ultramarines* is preserved also after substitution.

3°. By the replacement of Na by Ag, the constant a_0 of the fundamental grating is *diminished*.

4°. Notwithstanding all the analogies mentioned, the relative intensi-

ties of corresponding interference-images in the spectrograms of the *ultramarines* and of the *silver-ultramarines* are appreciably modified. (Fig. 1; to compare *A* and *B*).

Fig. 1.

§ **5.** The fact that the gradual replacement of *Na* in *ultramarine-blue* by *Ag* is really connected with a *gradual diminution* of the edge a_0 of the fundamental cell, could, moreover, also be proved in the following way.

If *ultramarine-blue* be digested during several days on the waterbath with an ammoniacal solution of *silverchloride,* then, as HEUMANN [1] stated

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[1] K. HEUMANN, loco cit., **201,** 282, (1880).

already, the Na of the original compound gets partially substituted by Ag. HEUMANN thought that in this way *two thirds* of the Na-atoms were replaced by Ag. In our experiments the original *ultramarine-blue* was heated during 6 days on the waterbath with an ammoniacal solution of *silver-chloride*, which was three times refreshed: the product obtained (N^0. 19ı) appeared yet to contain 39,4 $^0/_0$ Ag. This product was again treated in the same way during 10 subsequent days: now the carefully purified product N^0. 19ıı), gave on analysis: 31,6 $^0/_0$ Ag. A third treatment, however, gave again a product (N^0. 19ııı) with 34,2 $^0/_0$ Ag; evidently the substitution of Na by Ag does not stop at a limiting stoechiometrical proportion of both constituents, as HEUMANN believed, but the reaction goes on until a state of *equilibrium* is reached, — in the same way as occurs in treating the *permutites* with salt-solutions of varied concentrations.

The results of the analysis of the ROENTGEN-spectrograms obtained with these preparations, are collected in the Tables V and VI.

Moreover, a product was prepared by heating, during 50 hours in a flask with reflux-cooler, the original *silver-ultramarine* with a strong solution of 20 times its weight of *sodium-chloride*. This blueish coloured product (N^0. 9) still contained, besides sodium, 7,75 $^0/_0$ Ag; its ROENTGEN-spectrum gave the results collected in Table VII (Compare Fig. 1 F).

From the results of the analysis, it becomes evident, that besides the original *silver*-compound, there is nowhere present here a pure compound formed by substitution in simple stoechiometrical proportions:

For: $Ag_6Al_4Si_6O_{23}S_3$,   the *silver*-content is calculated at: 46.6$^0/_0$ Ag; obs.: 46.5$^0/_0$ Ag.   (3)

For: $Ag_5NaAl_4Si_6O_{23}S_3$,   „   „   : 41.3$^0/_0$ Ag; $\left\{ \right.$ obs.: 39.4$^0/_0$ Ag. (19ı)

For: $Ag_4Na(NH_4)Al_4Si_6O_{23}S_3$,   „   „   : 35.5$^0/_0$ Ag;

For: $Ag_3Na(NH_4)_2Al_4Si_6O_{23}S_3$,   „   „   : 28.7$^0/_0$ Ag; $\left.\right\}$ obs.: 31.6$^0/_0 Ag$. (19ıı)

For: $Ag_2Na(NH_4)_3Al_4Si_6O_{23}S_3$,   „   „   : 20.8$^0/_0$ Ag;   (19ııı)

For: $AgNa_5Al_4Si_6O_{23}S_3$ (in exp. N^0. 9), „   „   : 11.2$^0/_0$ Ag; obs.: 7.75$^0/_0$ Ag. (9)

Moreover, if the original *silver*-compound n^0. 3 be heated in a porcelain crucible with an excess of *sodium-iodide* just above the meltingpoint of this salt, and the molten mass be kept at this temperature for a longer time, then, after purification, an *ultramarine-blue* (n^0. 4) is formed, which, however, appears still to contain 6 $^0/_0$ Ag; its powder-spectrogram (see Table VIII) is almost identical with that of preparation n^0. 9, also with respect to the relative intensities of the successive diffraction-lines, and with that of the original GUIMET-*blue* itself.

If the extremely weak influence on the size of the grating-constant a_0, which is exerted, as we shall see later-on, by the substitution of Na by the (NH_4)-radicle, be now left aside for a moment, the constant a_0 of the preparations n^0. 3, 4, 9, 19ı, 19ıı, 19ııı, etc., appears to vary as follows:

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TABLE V. Powderspectrogram of Silver-sodium-Ultramarine No. 19¹.
(Contains: 39.4 %Ag; Fig. 1 C).

N⁰. of Image:	2l in m.m.:	λ:	Estim. intens.:	$\Sigma(h^2)$:	$\frac{\theta}{2}$:	$sin^2\frac{\theta}{2}$ (ob- served)	$sin^2\frac{\theta}{2}$ (calcu- lated):	Indices:
1	37.5	α	6	6	12°6′	0.0439	0.0439	(211)
2	43.7	β	3	10	14°6′	0.0593	0.0595	(310)
3	48.8	α	7	10	15°45′	0.0737	0.0732	(310)
4	53.5	α	5	12	17°16′	0.0881	0.0878	(222)
5	58.3	β	1	18	18°50′	0.1042	0.1071	(330) and (411)
6	58.9	α	5	14	19°0·	0.1059	0.1025	(321)
7	64.2	β	1	22	20°43′	0.1251	0.1310	(332)
8	65.7	α	6	18	21°13′	0.1310	0.1318	(330) and (411)
9	68.9	α	2	20	22°14′	0.1432	0.1464	(420)
10	74.0 *)	α	3	22	23°56′	0.1646	0.1610	(332)
11	·79.5	α	1·	26	25°39′	0.1874	0.1903	(431) and (510)
12	89.6	α	·1	32	28°54′	0.2336	0.2342	(440)
13	92.6	α	2	34	29°53′	0.2482	0.2489	(433) and (530)
14	95.5	α	1	36	30°52′	0.2622	0.2635	(600)
15	98.5	α	1	38	31°46′	0.2774	0.2782	(532) and (611)
16	100.8	α	1	40	32°31′	0.2889	0.2928	(620)
17	104.1	α	1	42	33°35′	0.3060	0.3074	(541)
18	107.0	α	2	44	34°31′	0.3211	0.3221	(622)
19	112.0	α	1	48	36°8′	0.3477	0.3514	(444)
20	115.5	α	1	50	37°16′	0.3667	0.3660	(543); (505); (710)
21	120.6	α	3	54	38°54′	0.3940	0.3953	(633) and (721)
22	126.3	α	1	58	40°44′	0.4258	0.4246	(730)
23	178.6	α	1	98	57°35′	0.7130	0.7125	(853)
24	208.0	α	2	116	67°6′	0.8486	0.8490	(864) and (10.04)

*) Only observed at one side of the central spot.
Radius of the camera: 44.4 mm. $V = 55000$ Volt; Time of exposure: 2 hours.
$\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U.; $a_0 = 9.00$ A.U.

Quadratic equation: $sin^2\frac{\theta}{2} = 0.00732\,(h^2 + k^2 + l^2)$, for the α-radiation.

$sin^2\frac{\theta}{2} = 0.00595\,(h^2 + k^2 + l^2)$, for the β-radiation.

TABLE VI. Powder-spectrogram of Silver-sodium-ammonium-Ultramarine No. 19^{II}.
(Analysis: 31,6 % Ag; Fig. 1 D).

No. of Image:	$2l$ in mm.	Estim. intensities:	$\Sigma(h^2)$:	λ:	$\dfrac{\theta}{2}$:	$sin^2\dfrac{\theta}{2}$ (ob-served):	$sin^2\dfrac{\theta}{2}$ (cal-culatated):	Indices:
1	37.7	6	6	α	12° 9′	0.0443	0.0439	(211)
2	44.2	2	10	β	14°15′	0.0605	0.0599	(310)
3	48.9	9	10	α	15°46′	0.0738	0.0732	(310)
4	53.7	4	12	α	17°19′	0.0886	0.0878	(222)
5	58.2	1	14	α	18°46′	0.1035	0.1025	(321)
6	59.8	2	18	β	19°17′	0.1090	0.1062	(330); (411)
7	66.2	6	18	α	21°21′	0.1325	0.1318	(330); (411)
8	69.1	1	20	α	22°17′	0.1438	0.1464	(420)
9	73.8	1	22	α	23°48′	0.1628	0.1610	(332)
10	80.0	2	26	α	25°48′	0.1894	0.1903	(431); (510)
11	83.1	1	34	β	26°48′	0.2033	0.2006	(433); (530)
12	90.0	3	32	α	29° 2′	0.2356	0.2342	(440)
13	93.1	3	34	α	30° 1′	0 2502	0.2490	(433); (530)
14	95.9	2	36	α	30°56′	0.2642	0.2635	(600)
15	99.1	2	38	α	31°58′	0.2803	0.2782	(611); (532)
16	100.5	1	40	κ	32°25′	0.2874	0.2928	(620)
17	104.5	1	42	α	33°42′	0.3078	0.3074	(541)
18	107.2	2	44	α	34°35′	0.3222	0.3221	(622)
19	112.5	1	48	α	36°17′	0.3502	0.3514	(444)
20	120.6	3	54	α	38°54′	0.3943	0.3953	(633); (721)
21	126.6	1	58	α	40°50′	0.4275	0.4246	(730)
22	172.5	1	94	α	55°39′	0.6816	0.6881	(763)
23	179.0	1	98	α	57°45′	0.7152	0.7174	(853)

Radius of the camera; 44.4 mm. $V = 55000$ Volt. Time of exposure: 3 hours.
$\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U.; $a_0 = 9.00$ A.U.

Quadratic equation: $sin^2\dfrac{\theta}{2} = 0.00732 \cdot (h^2 + k^2 + l^2)$, for the α-radiation.

$sin^2\dfrac{\theta}{2} = 0.00599 (h^2 + k^2 + l^2)$, for the β-radiation.

TABLE VII. Powderspectrogram of a Silver-sodium-Ultramarine N⁰. 9.
(From Silver-Ultramarine by heating with a solution of NaCl during 50 hours.)
Contains: 7.75 % Ag; Fig. 1 F.

N⁰. of Image:	$2l$ in mm.:	λ :	Estim. intensities:	$\Sigma(h^2)$:	$\dfrac{\theta}{2}$:	$sin^2\dfrac{\theta}{2}$ (ob-served):	$sin^2\dfrac{\theta}{2}$ (cal-culated)	Indices:
1	37.5	α	10	6	12°6′	0.0439	0.0436	(211)
2	43.7	β	1	10	14°6′	0.0593	0.0591	(310)
3	48.6	α	7	10	15°41′	0.0731	0.0727	(310)
4	53.2	α	7	12	17°10′	0.0871	0.0872	(222)
5	58.9	α	4	14	19°0′	0.1059	0.1018	(321)
6	61.6	α	2	16	19°52′	0.1155	0.1163	(400)
7	64.9 *)	β	1	22	20°56′	0.1278	0.1300	(332)
8	65.9	α	6	18	21°16′	0.1315	0.1309	(330) and (411)
9	68.7	α	1	20	22°14′	0.1426	0.1454	(420)
10	73.5	α	2	22	23°43′	0.1618	0.1599	(332)
11	80.2	α	5	26	25°52′	0.1903	0.1890	(431) and (510)
12	82.6	β	1	34	26°39′	0.2012	0.2009	(433) and (530)
13	86.4	α	2	30	27°52′	0.2185	0.2181	(521)
14	89.4	α	4	32	28°50′	0.2326	0.2326	(440)
15	92.4	α	2	34	29°49′	0.2473	0.2472	(433) and (530)
16	95.6	α	3	36	30°50′	0.2627	0.2617	(600)
17	98.3	α	2	38	31°43′	0.2764	0.2763	(532) and (611)
18	100.8	α	1	40	32°31′	0.2889	0.2908	(620)
19	104.5 *)	α	1	42	33°43′	0.2081	0.3053	(541)
20	107.0	α	4	44	34°31′	0.3211	0.3199	(622)
21	112.0	α	2	48	36°8′	0.3477	0.3490	(444)
22	115.0	α	1	50	37°6′	0.3639	0.3635	(543); (505); (710)
23	120.5	α	4	54	38°52′	0.3938	0.3926	(633) and (721)
24	145.9 *)	α	1	74	47°5′	0.5361	0.5380	(743); (750); (831)
25	149.4 *)	α	2	76	48°12′	0.5557	0.5525	(662)
26	151.8 *)	α	1	78	48°58′	0.5690	0.5671	(752)
27	154.0 *)	α	1	80	49°41′	0.5814	0.5816	(840)

*) Only observed at one side of the central spot.

Radius of the camera: 44.4 mm. $V = 55000$ Volt; Time of exposition: 2 hours.

$\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U.; $a_0 = 9.03$ A.U.

Quadratic equation: $sin^2\dfrac{\theta}{2} = 0.00727\ (h^2 + k^2 + l^2)$, for the α-radiation.

$$sin^2\dfrac{\theta}{2} = 0.00591\ (h^2 + k^2 + l^2),\ \text{for the}\ \beta\text{-radiation.}$$

If the *silver*-content is:

$$0 \quad \% \ Ag : a_0 = 9.06 \text{ A.U.}$$
$$7.75\% \ Ag : a_0 = 9.03 \text{ A.U.}$$
$$31.6\% \ Ag : a_0 = 9.00 \text{ A.U.}$$
$$39.4\% \ Ag : a_0 = 9.00 \text{ A.U.}$$
$$46.5\% \ Ag : a_0 = 8.96-8.99 \text{ A.U.}$$
$$51 \quad \% \ Ag : a_0 = 8.93 \text{ A.U.}$$

TABLE VIII. Powderspectrogram of *NaAg*-Ultramarine No. 4.
Contains about 6% *Ag*. (Fig. 1 G)

No. of Image:	$2l$ in mm.:	Estim. intensities:	λ:	$\|\Sigma(h^2)\|$:	$\dfrac{\theta}{2}$:	$sin^2\dfrac{\theta}{2}$ (observed):	$sin^2\dfrac{\theta}{2}$ (calculated):	Indices:
1	37.1	9	α	6	11°58′	0.0430	0.0431	(211)
2	47.9	6	α	10	15°27′	0.0710	0.0718	(310)
3	52.9	7	α	12	17° 4′	0.0861	0.0862	(222)
4	54.6	3	α	14	17°37′	0.0916	0.1004	(321)
5	57.4	1	β	18	18°31′	0.1010	0.1049	(330) and (411)
6	65.4	5	α	18	21° 6′	0.1296	0.1292	(330) and (411)
7	79.3	5	α	26	25°35′	0.1864	0.1867	(431) and (510)
8	81.0	1	β	34	26° 8′	0.1940	0.1982	(433) and (530)
9	85.7	1	α	30	27°39′	0.2153	0.2154	(521)
10	88.7	4	α	32	28°37′	0.2294	0.2298	(440)
11	91.5	3	α	34	29°31′	0.2427	0.2441	(433) and (530)
12	94.6	3	α	36	30°31′	0.2578	0.2585	(600)
13	97.4	3	α	38	31°25′	0.2717	0.2729	(532) and (611)
14	106.3	4	α	44	34°18′	0.3176	0.3159	(622)
15	108.1	1	α	46	34°53′	0.3272	0.3303	(631)
16	111.6	2	α	48	36° 0′	0.3455	0.3446	(444)
17	114.4	1	α	50	36°54′	0.3605	0.3590	(543);(505);(710)
18	119.2	3	α	54	38°27′	0.3867	0.3877	(633) and (721)

Radius of the camera: 44.4 mm. $V = 55000$ Volt; Time of exposure: 3 hours.
Wavelength: $\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U.; $a_0 = 9.09$ A.U.

Quadratic equation: $sin^2\dfrac{\theta}{2} = 0.00718 \ (h^2 + k^2 + l^2)$, for the α-radiation.

$$sin^2\dfrac{\theta}{2} = 0.00583 \ (h^2 + k^2 + l^2), \text{ for the } \beta\text{-radiation.}$$

From these data it becomes very evident that the constant a_0 has really a tendency to *decrease*, if the *silver*-content of the preparation *increases*.

Indeed, the atomic radius for *silver*, which differs, after all, only slightly from that of *sodium*, seems, in most cases, to be somewhat *smaller* than the last one. The influence of the substitution is strongest on the side of the pure *Na*-compound: a content of 6 $^0/_0$, or 8 $^0/_0$ Ag is in this respect not yet very appreciable, but becomes discernible only, if perhaps more than a 15 $^c/_0$ Ag is introduced. This may be seen e.g. from a graph, the abscissae of which are the *silver*-contents. The same phenomenon is observed with respect to the relative intensities. For a dozen interference-images common to both kinds of *ultramarines*, namely: (211); (310); (222); (321); (330), resp. (411); (431), resp. (510); (521); (440); (433), resp. (530); (532), resp. (611); (622); and (633), resp. (721), — the most important differences in this respect for the *silver*-compound in comparison with the *sodium*-compound are the following.

For the *Na*-compound $I_{(211)}$ is always *greater* than $I_{(310)}$; also $I_{(222)}$ always is *greater* than $I_{(310)}$; $I_{(330), (411)}$ is *smaller* than $I_{(322)}$; $I_{(521)}$ has yet a discernible size; $I_{(440)}$ is *greater* than $I_{(433), (530)}$; and the last-one is, although almost equal, also *greater* than $I_{(532), (611)}$; and $I_{(622)}$ is *greater* than $I_{(532), (611)}$ and also *greater* than $I_{(633), (721)}$. For the Ag-compound it is just the reverse in these respects.

From Fig. 1, in which on the horizontal axis the subsequent values of $(h^2 + k^2 + l^2)$ are graphically drawn, all this may easily be seen in comparing the figures 1^A, 1^B, 1^G; furthermore, in comparing the figures 1^C and 1^D for the *silver-sodium*-compounds, which have, — with the exception of a few images like (532) in 1^C and (433) in 1^D, — yet completely the character of Fig. 1^B. On the contrary, Fig. 1^F shows already the type of the *Na-ultramarines*, although the compound yet contains 8 $^0/_0$ Ag. The characteristic intensities of the *silver*-compound seem to become discernible, as soon as more than 12 or 15 $^0/_0$ Ag has taken the place of the equivalent quantity of *Na*.

§ 6. GUIMET [1] first demonstrated, that the *sulphur* in the *ultramarines* may be substituted by *selenium* or *tellurium*. Through kind intervention of M. A. GUILLOCHIN, director-general of the *Usines d'Outremer*-GUIMET in *Fleurieu s/Saône*, who put a small quantity of GUIMET's original, scarlet-red *selenium-ultramarine* at our disposal, — we were able to study this substance and to compare its properties with those of the corresponding *sulpher*-derivatives. The brilliantly red powder was purified and freed from the last traces of adhering *selenates* or *selenites* by repeatedly washing with distilled water; the final product (N^0. 80) was used in these experiments and partially converted into the corresponding *silver-selenium-*

[1] E. GUIMET, Bull. de la Soc. chim. Paris, **27**, 480, (1877); Ann. de Chim. et Phys., (5), **13**. 102, (1878). Here also we wish once more to express our sincere thanks to M. GUILLOCHIN.

No. of Image:	$2l$ in mm.:	λ:	Estim. intensities:	$\Sigma(h^2)$:	$\dfrac{\theta}{2}$:	$sin^2\dfrac{\theta}{2}$ (ob-served):	$sin^2\dfrac{\theta}{2}$ (cal-culated):	Indices:
								TABLE IX. Powder-Spectrogram of Selenium-Ultramarine-red-GUIMET (N⁰. 80).
1	37.4	α	10	6	12°4′	0.0437	0.0430	(211)
2	42.0	β	1	10	13°33′	0.0550	0.0582	(310)
3	48.0	α	4	10	15°29′	0.0712	0.0716	(310)
4	53.1	α	9	12	17°8′	0.0868	0.0859	(222)
5	58.0	β	1	18	18°43′	0.1030	0.1048	(330) ; (411)
6	61.4	α	3	16	19°48′	0.1148	0.1146	(400)
7	65.1	α	8	18	21°0′	0.1284	0.1289	(330) ; (411)
8	71.0	β	1	26	22°55′	0.1516	0.1513	(431) ; (510)
9	72.9	α	2	22	23°31′	0.1592	0.1576	(332)
10	76.3	α	2	24	23°36′	0.1733	0.1718	(422)
11	79.3	α	8	26	25°34′	0.1862	0.1862	(431) ; (510)
12	82.0	β	1	34	26°27′	0.1984	0.1979	(433) ; (530)
13	85.8	α	2	30	27°40′	0.2156	0.2148	(521)
14	88.6	α	4	32	28°34′	0.2286	0.2291	(440)
15	91.7	α	4	34	29°35′	0.2437	0.2434	(433) ; (530)
16	94.6	α	3	36	30°31′	0.2578	0.2578	(600)
17	97.6	α	3	38	31°29′	0.2727	0.2721	(532) ; (611)
18	105.7	α	6	44	34°6′	0.3143	0.3150	(622)
19	108.3	α	2	46	34°56′	0.3279	0.3294	(631)
20	111.1	α	3	48	35°50′	0.3427	0.3437	(444)
21	114.0	α	3	50	36°46′	0.3583	0.3580	(543) ; (505) ; (710)
22	119.1	α	5	54	38°25′	0.3861	0.3866	(633) ; (721)
23	121.3	α	1	56	39°7′	0.3981	0.3999	(642)
24	124.6	α	1	58	40°12′	0.4166	0.4153	(730)
25	129.2	α	1	62	41°40′	0.4419	0.4439	(732)
26	131.4	α	1	64	42°23′	0.4544	0.4582	(800)
27	135.1	α	1	66	43°35′	0.4753	0.4726	(554) ; (741) ; (811)
28	145.0	α	1	74	46°48′	0.5314	0.5298	(750) ; (743) ; (831)
29	147.4	α	2	76	47°33′	0.5444	0.5442	(662)
30	149.5	α	1	78	48°13′	0.5560	0.5585	(752)
31	152.5	α	2	80	49°12′	0.5730	0.5728	(840)
32	165.5	α	1	90	53°23′	0.6442	0.6444	(851)
33	171.0	α	1	94	55°10′	0.6737	0.6730	(763)
34	173.6	α	1	96	56°1′	0.6876	0.6873	(844)

Radius of the camera: 44.4 mm. $V = 55000$ Volt. Time of exposure: 3 hours. Wavelength: $\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U. $a_0 = 9.10$ A.U.

Quadratic equation: $sin^2\dfrac{\theta}{2} = 0.00716\,(h^2 + h^2 + l^2)$, for the α-radiation.

$$sin^2\dfrac{\theta}{2} = 0.00582\,(h^2 + k^2 + l^2),\ \text{for the } \beta\text{-radiation.}$$

TABLE X. Powderspectrogram of Silverselenium-Ultramarine.
(Analysis: 25.8 % Ag).

No. of Image:	$2l$ in mm.:	Estim. intensities:	$\Sigma(h^2)$:	λ:	$\frac{\theta}{2}$:	$sin^2\frac{\theta}{2}$ (observed):	$sin^2\frac{\theta}{2}$ (calculated):	Indices:
1	37.9	8	6	α	12°14′	0.0449	0.0445	(211)
2	43.9	1	10	β	14°10′	0.0600	0.0603	(310)
3	49.3	7	10	α	15°54′	0.0750	0.0742	(310)
4	54.1	5	12	α	17°25′	0.0896	0.0890	(222)
5	57.3	3	14	α	18°29′	0.1005	0.1039	(321)
6	66.7	7	18	α	21°31′	0.1345	0.1336	(330) and (411)
7	70.0	1	20	α	22°35′	0.1475	0.1484	(420)
8	74.2	1	22	α	23°56′	0.1646	0.1632	(332)
9	78.0	1	24	α	25° 7′	0.1801	0.1781	(422)
10	81.1	2	26	α	26°10′	0.1945	0.1930	(431) and (510)
11	83.7	1	34	β	27° 0′	0.2061	0.2050	(433) and (530)
12	90.7	3	32	α	29°15′	0.2387	0.2374	(440)
13	93.7	4	34	α	30°13′	0.2533	0.2523	(433) and (530)
14	96.6	2	36	α	31°10′	0.2678	0.2671	(600)
15	99.7	3	38	α	32° 9′	0.2832	0.2820	(532) and (611)
16	102.0	1	40	α	32°55′	0.2953	0.2968	(620)
17	105.5	1	42	α	34° 2′	0.3132	0.3116	(541)
18	108.3	2	44	α	34°56′	0.3279	0.3265	(622)
19	110.6	1	46	α	35°41′	0.3403	0.3413	(631)
20	113.6	1	48	α	36°39′	0.3563	0.3562	(444)
21	116.5	1	50	α	37°35′	0.3720	0.3710	(543);(505);(710)
22	122.1	4	54	α	39°23′	0.4026	0.4007	(633) and (721)
23	127.3	2	58	α	41° 4′	0.4316	0.4304	(730)
24	132.7	3	62	α	42°48′	0.4616	0.4600	(732)
25	137.9	1	66	α	44°29′	0.4910	0.4897	(554);(741);(811)
26	140.6	1	68	α	45°21′	0.5061	0.5046	(644) and (820)
27	150.9	2	76	α	48°41′	0.5641	0.5639	(662)
28	156.7	2	80	α	50°33′	0.5963	0.5936	(840)
29	159.2	1	82	α	51°21′	0.6099	0.6084	(833)
30	164.5	1	86	α	53° 4′	0.6389	0.6381	(655)
31	169.8	1	90	α	54°47′	0.6675	0.6678	(851)
32	193.1	1	106	α	62°18′	0.7839	0.7865	(950)
33	197.2	2	108	α	63°37′	0.8025	0.8014	(666)
34	200.1	1	110	α	64°33′	0.8153	0.8162	(10. 3. 1)
35	206.9	1	114	α	66°45′	0.8442	0.8459	(774) and (855)
36	211.1	1	116	α	68° 6′	0.8609	0.8607	(864) and (10.0.4)

Radius of the camera: 44.4 mm. $V = 55000$ Volt. Time of exposure: 2 hours.
$\lambda_\alpha = 1.540$ A.U.; $\lambda_\beta = 1.388$ A.U.; $a_0 = 8.94$ A.U.

Quadratic equation: $sin^2\frac{\theta}{2} = 0.00742\ (h^2 + k^2 + l^2)$, for the α-radiation.

and $sin^2\frac{\theta}{2} = 0.00603\ (h^2 + k^2 + l^2)$, tor the β-radiation.

ultramarine (N⁰. 81) (with 25,8 % *Ag*, according to its analysis), whose colour is orange-yellow or brownish-yellow in this case, and which preparation was purified by frequent decantation and sedimentation. Of both these substances, which are poor in *Ag*-content, powder-spectrograms were made; the results of their analysis are collected in the Tables IX and X.
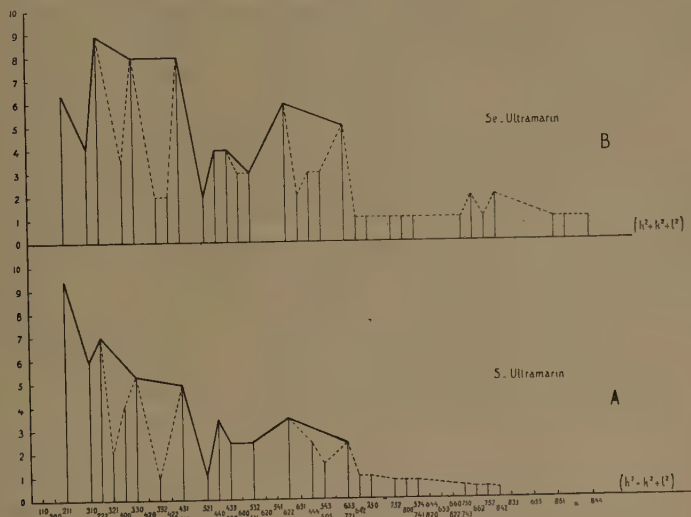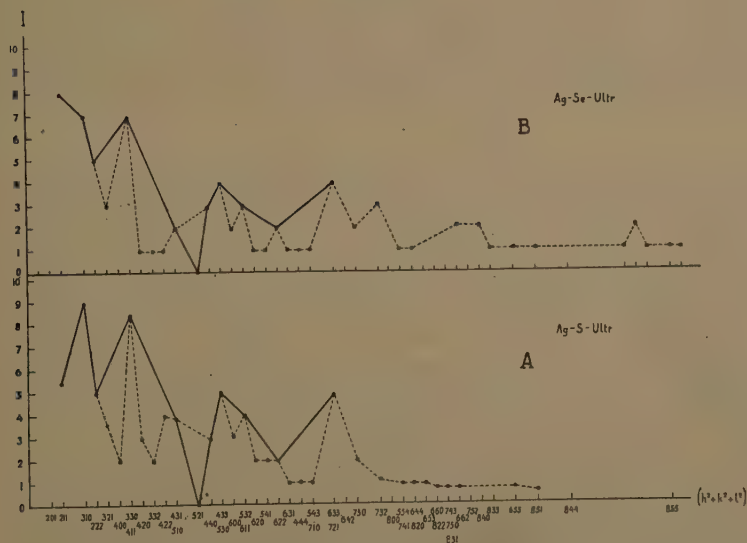

Fig. 2.

Fig. 3.

With the same time of exposure, viz. 3 hours, the background of the photographic film in the case of *selenium-ultramarine* appeared to be appreciably more and very homogeneously clouded, than in the case of the ordinary *sulphur-ultramarines*. The intensity of the diffraction-images was great and the different lines were very sharp.

From the data thus obtained, as well on comparing Fig. 2 and Fig. 3, — which in the same way as Fig. 1, give the estimated, relative intensities of corresponding diffraction-images in both patterns, — it may be deduced, that:

1stly. The substitution of S in the ordinary *ultramarines* by Se leaves the general structure unchanged. Under comparable conditions of exposure the film is, in the case of the *selenium*-derivatives, more intensily and generally clouded: which doubtlessly is caused by the greater diffracting power of the Se-atoms, in comparison with that of the S-atoms. However, the general character of the relative intensities of corresponding diffraction-images and their sequence, appears to remain the same; only a few exceptions are present, e.g. that $I_{(222)}$ in the case of the Se-compound is *greater* than $I_{(211)}$; and that the same occurs for $I_{(433),\,(530)}$ in comparison with $I_{(522),\,(611)}$ and for $I_{(422)}$, which last intensity in the case of the *S-ultramarine* is practically equal to zero. The grating-constant a_0 of the Se-derivative $(a_0 = 9,10 \, \text{A.U.})$ is only *very slightly* greater than that of *S-ultramarine*.

2ndly. The substitution of S by Se in *silver-ultramarine* does neither change its structure to any appreciable extent, while also the relative intensities of the successive corresponding images appear to be quite analogous in both cases, with the only exception (Fig. 3) that $I_{(211)}$ of the *Se-Ag-ultramarine* is *greater* than $I_{(310)}$, and the reverse occurs in the *S-Ag*-derivative. Also in this case both grating-constants a_0 do not differ appreciably.

§ 7. From the investigations described it must become clear now, that the substitution of the atoms of the alkali-metal in GUIMET-*blue* by more than about $15\,\%$ Ag, has an appreciably greater influence on the relative intensity-sequence of the diffraction-images in the corresponding ROENTGEN-spectograms, than has the substitution of the S-atoms in it by Se; this last substitution hardly, and only in some few respects, influencing the said intensities for some of the diffracting sets of planes, but chiefly causing only a general clouding of the background of the photographic films. In this an indication might be found that the Na-and Ag-atoms take a more important part in the formation of the "rigid" periodical structure of the *ultramarines*, than do the S- or Se-atoms. Notwithstanding this, it cannot yet be considered to be finally proved, that the S- (or Se)-atoms are, therefore, to be supposed to be statistically dispersed throughout the whole fixed structure, while the Na- (or Ag)-atoms would, at least partially, represent the true building-

stones in the fixed, invariable, periodical structure of these remarkable substances. The experience hitherto obtained can only give a few more arguments in favour of the probability of such a conception, but as yet no complete certainty about it. Further experiments in this direction have already been started.

§ 8. In the course of these investigations again a second tentative was made to find amongst the cubic zeolites a type of compound showing the peculiarities of the ROENTGENspectrograms, as observed in *hauyne, noseane* and the *ultramarines*. In this case *analcite* was chosen as the object of such a study; but also this beautifully crystallized and cubic *sodium-aluminium-silicate*, chemically closely related to the said silicates, appeared to possess *quite another structure* than the *ultramarines*. Although we intend to publish the more detailed data about the structure of *analcite* in the near future, — when at the same time we shall have an opportunity to discuss the question of the presence of "errant" constituents, namely of H_2O-molecules; in it, — we wish to communicate here already shortly some of the principal results of this investigation.

At our disposal were splendidly built, translucent crystals from the Cyclopic Isles, which were bordered principally by the forms {100} and {211}.

With the aid of powder-spectrograms, of BRAGG's spectrograms on the faces of {100} and {211}, and of rotation-spectrograms in which the crystals were turned round the directions [100], [110] and [111], — we were able to demonstrate, that this mineral has a structure to be described by the space-group O_h^{10}. In each cubic cell, the edge of which is equal to 13.71 A.U., 16 molecules are present of the composition: $NaAl\,Si_2\,O_6, H_2O$. If to the 16 Na-, and the 16 Al-atoms the two only possible, parameterless 16-fold places be attributed:

$[0\ 0\ 0]$; $[\frac{1}{4}\ \frac{1}{4}\ \frac{1}{4}]$; $[\frac{3}{4}\ \frac{3}{4}\ \frac{3}{4}]$; $[\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2}]$; $[\frac{1}{8}\ \frac{1}{8}\ \frac{1}{8}]$; $[\frac{1}{8}\ \frac{7}{8}\ \frac{3}{8}]$; $[\frac{3}{8}\ \frac{1}{8}\ \frac{7}{8}]$; $[\frac{7}{8}\ \frac{3}{8}\ \frac{1}{8}]$;

$[\frac{1}{2}\ \frac{1}{2}\ 0]$; $[\frac{3}{4}\ \frac{3}{4}\ \frac{1}{4}]$; $[\frac{1}{4}\ \frac{1}{4}\ \frac{3}{4}]$; $[0\ 0\ \frac{1}{2}]$; $[\frac{7}{8}\ \frac{7}{8}\ \frac{7}{8}]$; $[\frac{3}{8}\ \frac{1}{8}\ \frac{5}{8}]$; $[\frac{5}{8}\ \frac{7}{8}\ \frac{1}{8}]$; $[\frac{1}{8}\ \frac{5}{8}\ \frac{7}{8}]$;

$[\frac{1}{2}\ 0\ \frac{1}{2}]$; $[\frac{3}{4}\ \frac{1}{4}\ \frac{3}{4}]$; etc. | $[\frac{3}{8}\ \frac{3}{8}\ \frac{3}{8}]$; $[\frac{1}{8}\ \frac{3}{8}\ \frac{5}{8}]$; etc.

to one of both kinds of atoms, | to the other of both kinds of atoms,

and if to the Si-atoms the 32-fold position 32 f (after WIJCKOFF's notation; it has *one* variable parameter u) be given, — then there are yet available only two 24-fold, parameterless positions, two 48-fold positions with a single variable parameter v, and one 96-fold position with three variable parameters. Indeed, in this way it appears possible also to fix the positions of the 96 O-atoms of the complex $Na_{16}\,Al_{16}\,Si_{32}\,O_{96} + + H_{32}\,O_{16}$; but *there is no place any more available for the* 16 O-atoms *of the water-molecules*, and it seems inevitable, to assume these very

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"mobile" crystalwater-molecules to be also "errant" constituents of the structure. In short we hope to resume this topic in detail. As formerly already pointed out, for the *ultramarines* the space-group O_h^{10} cannot be taken into account.

Groningen, Laboratory for Inorganic and Physical
Chemistry of the University.

Pathology. — *On the isolation of a substance with carcinolytic properties from the reticulo-endothelial system* [1]). By N. WATERMAN and L. DE KROMME. (Communicated by Prof. G. VAN RIJNBERK.)

In an earlier memoir we demonstrated what FREUND and KAMINER had previously pointed out, namely that the normal blood-serum and the normal skin contain a substance capable of dissolving cancer-cells in vitro.

This property diminishes, or is totally lost, when a carcinomatous process arises.

Experimental research has opened up the possibility of studying the changes in this property on the dorsal skin of the tarred white mouse.

It has now been found the smearing with tar actually diminishes this lytic propertiy of the dorsal skin of the mouse, and that this property disappears altogether on the appearance of the carcinoma.

Further, the influence of Röntgen-rays upon this property appeared to be of great importance. If, for instance, skin-extracts possessed of a certain lytic power are subjected to Röntgen-rays, this power may be increased or diminished, according to the *length of time of the irradiation* (180 K.V., 2 m.A., 25 cm. unfiltered).

Inversely, a non-lytic extract from a carcinomatous skin may be made lytic again by röntgenization, and the same oscillations in the lytic action will be dependent upon the duration of the irradiation.

That this lytic power must have something to do with the disappearance of tumor-cells under the influence of irradiation, is evident from the fact that no destruction of the cells by the rays takes place in an inorganic environment, but does, on the contrary, occur when they are found in an originally non-lytic carcinomatous serum or organ-extract. (Biochem. Zeitschr. Vol. **182**, p. 377, and Fortschr. a. d. Geb. der Röntgenstrahlen, Vol. **35**, part. 4).

Whereas our results so far are more or less in agreement with those of FREUND and KAMINER, we differ from them widely on some points.

In particular their view that the lytic substance must be considered to be a certain saturated dicarbonic acid, we were unable to substantiate.

Neither succinic nor suberic acid, both of which may be taken as representatives of the above type of acids (as indicated by FREUND and KAMINER), exhibited any lytic action in our experiments.

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[1]) After researches carried out in the Anthonie van Leeuwenhoekhuis at Amsterdam.

We are of opinion that the researchers in Vienna have been led astray by by-products.

We therefore felt constrained to take in hand once more the isolation of the lytic substance, and to do so after our own method.

Our hypothesis was that in the organism there must be a system which governs the generation and the regeneration of all the cells of the body, and which must be specially active wherever strong regenerative processes are continually at work.

We, therefore, determined in which organs the lytic element is present in the greatest quantity, and endeavoured to find a method of preparation whereby that element may be isolated from the organs. The organs examined were the liver, spleen, pancreas, thymus, adrenals and lymphatic glands.

Previous researches by FREUND and KAMINER had demonstrated that the active matter must be soluble in ether, and this was confirmed by us. But the experience with modern hormone preparations does not appear to warrant the conclusion that the substance has a lipoïd character.

Now, while the ether extracts of almost all organs were found to possess a more or less lytic action, the extracts of the lymphatic glands, spleen and thymus possessed this in a much higher degree than that of the other organs. This was in accordance with the views of various researchers who have attributed the power of resistance to carcinoma in particular to the quality of the connective tissue and to the reticulo-endothelial and lymphocytic apparatus.

First of all, however, the method of extraction had to be improved.

An extraction of calves' spleens and lymphatic glands was made with 10 to 15 times the quantity of ether under return-flow cooling for 8 hours. The ether extract was then partially evaporated and the last remains of the ether driven off by the air-current. The watery residue was treated with an excess of aceton. This caused a white flocculent precipitate, which was washed several times with aceton and finally dried on the filter. This dried precipitate was then rubbed down with 96 % alcohol (or methyl-alcohol), whereby the greater part is dissolved. After being filtered, however, a whitish-grey substance is retained, which is separated from the last traces of alcohol in the filter at 37 %. This whitish-grey powder is finally shaken briskly with a phosphate-buffer in phys. NaCl solution (1 part buffer-solution after SÖRENSEN of P_H 7.7 with 5 parts phys. NaCl solution). Hereby the active matter is dissolved.

In this way an aceton-precipitate of several hundreds of milligrammes is obtained from 500 Gr. of organ. After washing with alcohol, only fractions of one milligramme pass into the watery solution. This preparation, however, must be considered as still very impure. As a rule, a clear solution of the active matter can be obtained by this method. The presence of any traces of lipoïds hinders the determination of the lytic power. The solution

contains no ferment (trypsine: Fuld-Gross, or lipase: tributyrine test according to the stalagmometric method) and is devoid of albumen.

In determining the lytic power, account should be taken of the P_H of the environment in which this determination takes place. It appeared that an acid reaction counteracts the lysis; an alkaline (P_H of blood serum), on the other hand, promotes it. We therefore carried out all our determinations with a P_H of 7.1.

By this method a substance can be obtained from the spleen and lymphatic glands of the calf, which is able to dissolve cancer cells. The lytic power thus far has been brought up to 81 %. Further purification, which is very necessary, will probably raise this percentage still higher.

The question now was to see whether this artifically prepared extract would also be affected by the X-rays in the same way as serum or skin extract. This proved to be the case.

EXAMPLE:

Extract of lymphatic glands	Lytic power in $\%$
Non-radiated	81
After 1 hour's irradiation	61
„ 2 hours' „	46
„ 4 „ „	62

SUMMARY.

1. Normal blood-serum and normal organs contain a substance capable of dissolving cancer-cells.

This substance is influenced by Röntgen-rays.

Extracts of carcinomatous organs lack this power, but obtain it by irradiation.

2. We have succeeded in isolating this substance from the reticulo-endothelial system, but as yet only in an impure state.

The solutions obtained contain neither tryptic nor lipatic ferment, and are free from albumen.

The lytic power of these solutions changes under the influence of Röntgen-rays.

Botany. — *Rapid flowering of Early Tulips ("Van der Neer").* (Communication N⁰. 24 of the Laboratory for Plant-physiological Research, Wageningen, Holland.) By Miss IDA LUYTEN. (Communicated by Prof. A. H. BLAAUW.)

§ 1. *Introduction.*

In connection with the research of BLAAUW (1926) on the rapid bringing into bloom of Darwin-tulips, this year experiments were made on the early forcing of "Early Tulips". The influence of the various temperature-combinations in summer having only been traced on the Darwin-tulip Pride of Haarlem (BLAAUW and VERSLUYS 1925, LUYTEN, JOUSTRA and BLAAUW 1925, MULDER and BLAAUW 1925), the plans for the "Early tulips" were based on the results of these papers combined with the experience gained by BLAAUW (1926) with the rapid flowering of these tulips, in the hopes that the early tulip would not differ too much from the late Darwin-tulip.

It has appeared to us, that the Single Early Tulip *Van der Neer* comes very near to the Darwin-tulip in many respects ; from the researches it also followed however, that among varieties of the "Single Early Tulip" as *Proserpine, La Remarquable* and *Van der Neer* there exist great differences in response to the external factors examined, such as : the various temperatures and light and dark. With respect to the "Early Tulips" therefore we cannot draw up one definite scheme ; to the definite varieties separate rules apply. Subjoined we give the results obtained in forcing the variety *Van der Neer*.

§ 2. *The temperature-treatment during the summer till planting-time.*

As summer-treatment of the Darwin-tulip for early flowering 3 weeks 20° + 8 weeks 9° was chosen by BLAAUW (1926). Indeed it had appeared at the time (communications 17, 18, 19), that with respect to the rate of forming the flower 17° was as favorable as 20°, but 17° gave an increase of the number of floral parts and transitions between tepals and foliage-leaves. Especially this latter always increases the chance of the flower growing deformed or crooked.

This year however for *Van der Neer* a temperature of 17° was used by the side of 20°, in order to trace whether this offered a more favourable combination with 9° with regard to flowering earlier.

On *June 25* the 220 tulips arrived at our laboratory after lifting straight from the field. The tulips were divided *into equal numbers and weights,* so that we might be sure afterwards, that every part of the research was based on the same initial weight. We placed 8×12 individuals at $17°$ C., whilst 8×12 and 1×18 were put at $20°$ C. Every group of 12 individuals weighed 405 grams; the 18 had $1\frac{1}{2} \times$ that weight. At the same time on June 25th 10 bulbs, weighing consequently $337\frac{1}{2}$ grs., were opened in order to trace how the condition was at that moment. It appeared that all bulbs were in *Stage I,* i.e. that the terminal vegetation-point was still splitting off foliage-leaves (Table I), 2 to 3 foliage-leaves had been formed per bulb. As the table shows, the length of the outer foliage-leaf was measured; this is still very small on this date, viz. averagely 1.15 mms. $\pm$ 0.016. As already fully described in previous publications, staining in an aqueous solution of iodine and iodide of potassium is required to clearly see all the parts split off by the vegetation-point. For an accurate description of the stages we refer to MULDER (1927).

We wanted to transfer the tulips to $9°$, when the flower would have reached *Stage III,* i.e. when the first whorl of tepals would be found as individual protuberances. Every time we wanted to state this, bulbs had to be opened to judge of the floral growing-point; for this control 2×12 bulbs had been reserved.

From the Tables I and II we see, that the bulbs were not opened until July 13; in $17°$ the terminal vegetation-point of 2 bulbs was respectively in stage II, i.e. the vegetation-point was raising itself, so it was passing on to flower-formation, and in stage II à III, i.e. there was a beginning of the first whorl of tepals. In $20°$ both bulbs were in stage II. The opened bulbs having not yet reached the desired stage, the opening of 2 bulbs from each temperature was deemed sufficient. On *July 16,* 5 bulbs from $17°$ (Table I) all appear to have reached the desired stages III or III+; all sets from $17°$ therefore, are then transferred to $9°$.

On this date the bulbs from $20°$ appeared to have progressed no farther than stages II and II — (Table II); on *July 20* however these groups could also be transferred to $9°$, since by that time 4 opened bulbs showed stages which had passed III. Probably these bulbs had already attained stage III on July 19.

$4\frac{1}{2}$ weeks after being transferred to $9°$, the bulbs were *planted,* whilst on those days respectively *Aug. 16* and *Aug. 20* (Tables I and II) groups of 11 and 10 bulbs of both sets were controlled with regard to the fact whether the flower had really been finished in this period: indeed in both groups the flower appeared to be quite finished, except one flower of the exposure $17°$—$9°$, which was in stage VI+. At the same time some measures were taken, which we can read from the tables.

The tables moreover show: 1⁰. that the successive flower-stages are reached in a very rapid tempo, from which it follows, that the control-days should follow very closely; 2⁰. that the length of the outer foliage-

TABLE I. Condition from the lifting to the planting.
Exposure: 21 days 17° C. $+$ 4^1/$_2$ weeks 9° C.

Date	Length outer foliage-leaf in mms.		Number of foliage-leaves per 10 bulbs	Stage	Length .apex of flower to base of stem in mms.	
	M	m			M	m
June 25, 1926 Beginning of the experiments	**1.15** $\pm$ 0.016	n = 10	24	I n = 10		
July 13	2. — 2.25			II and II to III	—	
July 16 transferred to 9°	**2.61** $\pm$ 0.12	n = 5		III and III+	—	
Aug. 16, 1926 Day of planting	**10.18** $\pm$ 0.42	n = 11	42		**6.16** $\pm$ 0.22	n = 11

TABLE II. Condition from the lifting to the planting.
Exposure: 25 days 20° C. $+$ 4^1/$_2$ weeks 9° C.

Date	Length outer foliage-leaf in mms.		Number of foliage-leaves	Stage	Length apex of flower to base of stem in mms.	
	M	m			M	m
June 25, 1926 Beginning of the experiments	**1.15** $\pm$ 0.016	n = 10	24	I n = 10		
July 13	1.55 1.55			II	—	
July 16	1.65 1.75			II and II —	—	
July 20 transferred to 9°	**2.28** $\pm$ 0.08	n = 4		IV; V—; VI+; VI — VII.	—	
Aug. 20, 1926 Day of planting	**10.46** $\pm$ 0.23	n = 10	40		**7.02** $\pm$ 0.26	n = 10

leaf on the day of transferring to 9° is greater in the group from 17° (Table I, July 16, 2.61 mms.) than in the group from 20° (Table II. July 20, 2.28 mms.), but that the bulbs with the preliminary treatment of 20° have caught up on the day of planting (Aug. 20) and now show an outer foliage-leaf about equally long as in the group 17°—9° on Aug. 16. The average length from the apex of the flower to the base of the stem is even somewhat greater now in 20° than in 17°.

§ 3. *From the planting to the transfer to the hothouses.*

All tulips were stripped of their outer, dry brown scale before the planting ; this happened to prevent the tulips as much as possible from raising themselves during the shooting of the roots. Moreover the axillary buds were removed from this brown scale. As plant-boxes we used boxes measuring 22 × 20 cms. clear diameter and having a depth of 18 cms.

Since BLAAUW (1926) in the early flowering of the Darwin-tulips was troubled with the drying out and also with the tipping of the stems and we did not know whether these phenomena would occur in this early tulip and whether they might be due to an excess of humidity, certain precautions were taken in these experiments in order to trace this.

For this purpose the group of 18 tulips with the preliminary treatment 20°—9° (we shall indicate the groups which were in the thermostate in 20° C. and next in 9° C. in this way, whilst we shall designate the groups from the other temperature-exposure 17°—9°) was planted into 3 boxes, each having 10 air-holes in the side-walls. In that way the Darwin tulips had been planted ; they had been liberally watered on the surface-soil. This group would be treated similarly. All other groups from 20°—9° and from 17°—9° were planted into boxes, the wood of which was entirely coated with paraffin : partly to prevent desiccation partly to preclude a possible influence of the wood. No holes were made in the side-walls, but 7 holes in the bottom. These boxes would never be watered on top, but were given a certain quantity at set times by immersing them in a cistern of water. The bottom of all the boxes was covered with a layer of fine gravel 1 cm. thick, whilst dune-sand was chosen as soil. Groups of six bulbs were placed in a box, the root-wall at 10 cms. below the edge of the box ; the box was filled in with dune-soil up to 2 cms. above the tip of the bulb. For the present, therefore, little soil was added, to facilitate measuring the "noses" ; afterwards when the plants were flowering the boxes were filled up to 4 cms. above the tip of the bulb.

Each group of 12 therefore was divided over two boxes. Accordingly we planted 6 groups each of 12 bulbs with a preliminary treatment of 17°—9° ; 6 groups of 12 bulbs each, and 1 group of 18 bulbs having had a preliminary treatment of 20°—9°. How the groups were divided afterwards, we shall discuss in § 4 in connection with their removal to the hothouses.

Now that the tulips have been planted, they are still kept at 9° ; but as they are now in a *room* which is being cooled, the temperature fluctuates more, that is between 8.5° and 10° C. *in the ground.* The soil in the boxes is covered with thick moist filter-paper to prevent it from drying out. The soil of the boxes coated with paraffin remains much moister than of the non-paraffined.

On *November 11* the average length of the "nose", i.e. that part of the foliage-leaves which shows from the bulb and is measured from the apex of the bulb to the apex of the outer leaf appeared to be 4.63 cms. $\pm$ 0.14 in the 71 bulbs 17°—9° C. (Table III). 72 bulbs had been planted ; one however had to be left on account of disease. This whole set 17°—9° C. was transferred to the hothouses, i.e. to the higher temperatures on this date. For the early flowering of the Darwin-tulip BLAAUW (1926) found $\pm$ 6 cms. as most desirable length for removal. Since the plant of the early tulip however remains much smaller than that of the Darwin-tulip, an *average of 4.5 cms.* was chosen in this case as average length of the "nose" as a standard for removal to the hothouses, that is to higher temperatures.

The "noses" of the groups 20°—9° were not yet so long on November 11 ; the average length (n = 71) was 3.44 cms. $\pm$ 0.10 on this date (Table IV). Not until *November 18* the average length was 4.59 cms. $\pm$ 0.12, so that by that time they too could be transferred to the higher temperatures in the hothouses.

We call attention to the fact that the start *the group 17°—9°* had in its development of 20°—9° and which amounted to 4 days on July 16 (cf. Tables I and II), has now increased to *7 days.*

§ 4. *The effect of the various temperatures and of light and dark on the rapid flowering.*

The tulips in the paraffined boxes were transferred to 3 different temperatures in the hothouses, viz. 22°—23° C., 19½°—20½° C. and 16°—17° C. Accordingly in every hothouse there were put two groups of each temperature-treatment on the tablets. These two groups were again divided by placing one group in the dark for the present, the other directly in the light. Our intention was to trace whether a temporal darkening affected the stretching and the rapid coming into bloom of the tulips. For this purpose black paper coverings were put on the two boxes of the one group ; they were 25 cms. higher than the boxes, so that the plants had room and plenty of air to grow. The 18 bulbs of the treatment 20°—9° planted into 3 non-paraffined boxes, were placed in a hothouse kept at 19½°—20½° C. As contrasted with the 3 other hothouses which were kept dry and the moisture of which fluctuated between 35 %—50 %, this hothouse was frequently syringed in consequence of which the moisture was kept at 75 %. The boxes in the moist hothouse were watered on the surface every day, whilst as already mentioned in § 3, to the boxes in the

other hothouses such a quantity of water was given by immersing them, that in these dry hothouses the moisture of the soil was the same in all boxes and much less than in the moist hothouse. In this system of immersing, in which a definite period was fixed for every hothouse, the covers need not be taken from the boxes and consequently the dark was not interrupted. From the tables III and IV we can read the division once more. Let us now first discuss the groups which have had

a preliminary treatment with 17°—9° C.

and which were transferred to the hothouses on November 11, 1926. Through the influence of the different hothouse-temperatures we soon see a difference in length in the groups which are in the light. In the groups in the dark we cannot state this for the present, since it is not allowed to take off the covers. In the groups in the light, the nose-length is greatest in the hottest house. On our measuring the nose-length in these groups on *November 22*, it appears that besides this outer foliage-leaf part of the stem found under this leaf has also been pushed from the bulb. From table III we see, that the length of these parts together averagely amounts to 19.18 cms. ± 0.91 (group B) in 22°—23° C., in 19½°—20½° averagely 15.89 cms. ± 0.52 (group D), and in 17°—18° it is not more than 13.03 cms. ± 0.61 (group F). Fig. 3 shows half of group B, that is from hothouse 22°—23°, whereas the box of Fig. 1 clearly shows, that the group from hothouse 19½°—20½° is less advanced ; the foliage-leaves are still rolled up in this case.

On Nov. 22 when we measured the groups which were in the light, we took the covers from the groups kept in the dark for the first time. It appeared, that the period of 11 days during which they had been in the dark, had already been too long, for practically all groups were etiolated ; and the groups kept in the higher temperatures, showed this most. Fig. 2 pictures to us what the group C from the hothouses 19½°—20½° C. looked like ; it likewise enables us to compare it with the group kept in the light in the same hothouse (Fig. 1). It appears that darkening especially promotes the vigorous growth of the part of the stem under the outer foliage-leaf. On comparing the average lengths of the outer foliage-leaves in Table III, we see, that they are fairly identical ; the averages of the parts of the stem under this leaf however show marked differences between the groups 'light' and 'dark', viz. between *A* and *B* 7.08 cms., between C and D 7.28 cms. and between *E* and *F* 6.07 cms. The long parts of the stem were no more erect, but nearly all of them were bent and crooked, which is clearly visible in the box from the dark in Fig. 2; this box may also be compared with the group *D*, placed in the *light* in the same hothouse (Fig. 1).

An other disadvantage of darkening is, that in all groups the leaves had remained firmly closed, while the groups in the light had started unfolding

TABLE III. The temperature-exposure 17° — 9°.

Group	Nov. 11 Average length of nose in cms.		Division		Nov. 22 Average lenght of part of stem + outer foliage leaf in cms.		Nov. 22 Average length of part of stem in cms.		Nov. 22 Average length of outer foliage leaf in cms.	Come into bloom	Number of flowers
	M	m	Temperatures	Light and dark	M	m	M	m			
A	3.67	± 0.15	22° −23°	dark	22.8	± 0.68	10.49 ± 0.42		12.40	Dec. 3	3 in bloom, 5 withered, 3 wh.-edged,
B	5.62	± 0.50	22° −23°	light	19.18	± 0.91	3.41 ± 0.45		15.77	Dec. 2	11 „ „ 1 1 bulb diseased
C	4.75	± 0.25	19.5°—20.5°	dark	22.82	± 0.82	9.22 ± 0.53		13.60	Dec. 3	5 „ „ 6 .. 1 „ „
D	4.33	± 0.23	19.5°—20.5°	light	15.89	± 0.52	1.94 ± 0.26		13.95	Dec. 3	10 „ „ 1 .. 1 „ „
E	5.25	± 0.22	17° −18°	dark	19.92	± 0.55	6.92 ± 0.34		13.0	Dec. 9	11 „ „ 1 „ „
F	4.14	± 0.24	17° −18°	light	13.03	± 0.61	0.85 ± 0.19		12.18	Dec. 9	10 „ „ 2 „ „

TABLE IV. The temperature-exposure 20° — 9°.

Group	Nov. 11 Average length of nose in cms.		Nov. 18 Average length of nose in cms.		Division		Nov. 22 Average length of nose in cms.		Come into bloom	Number of flowers
	M	m	M	m	Temperatures	Light and dark	M	m		
A	3.40	± 0.18	4.27	± 0.20	22° −23°	dark	9.33	± 0.36	Dec. 9	10 in bloom, 2 white-edged.
B	3.48	± 0.24	4.80	± 0.33	22° −23°	light	9.15	± 0.33	Dec. 7	12 „ „
C	3.32	± 0.10	4.85	± 0.52	19.5°—20.5°	dark	9.53	± 0.82	Dec. 13	11 „ 1 coloured
D	3.69	± 0.14	4.72	± 0.18	19.5°—20.5°	light	8.52	± 0.31	Dec. 9	10 „ 1 wh.-edged. 1 bulb diseased
E	3.32	± 0.20	4.30	± 0.23	17° −18°	dark	7.07	± 0.36	Dec. 13	12 „
F	3.52	± 0.10	4.64	± 0.18	17° −18°	light	7.59	± 0.25	Dec. 13	11 „ 1 withered
Non pa-raffined	3.02	± 0.22	3.95	± 0.25	19.5°—20.5°	light	—		Dec. 9	11 „ 1 „ 5 white-edged

FIG.1 23NOV.

O 17º 9º 20° LICHT

FIG.2 23NOV.

C 17º 9º 20° DONKER

FIG.3 23NOV.

B 17º 9º 22½° LICHT

FIG.4 23NOV.

B 20º 9º 22½° LICHT

FIG. 5 30NOV.

B 20º 9º 22½° LICHT

on reaching a sufficient length. So we see, that in the group *B* from 22°—23°, when the part of the stem between the tip of the bulb and the bottom-leaf is $3\frac{1}{2}$ cms., the outer foliage-leaf has already unfolded, so that the second foliage-leaf is usually revealed along its entire length (Fig. 3), while the group *D* from hothouse $19\frac{1}{2}$°—$20\frac{1}{2}$° (Fig. 1) starts unfolding when the part of the stem mentioned above average 1.94 cms., so that some apexes of the second foliage-leaves are already visible. In fig. 2 we notice that the foliage-leaves of the tulips kept in the dark still firmly keep together, in spite of the basal part of the stem averaging 10.5 cms.

From this moment all groups of the temperature-exposure 17°—9° are put in the light. The groups which had been in the dark, grow green and the foliage-leaves are going to unfold.

On *Nov. 25* some flowers of *the group B 22°—23°light* (Table III) are going to colour, on Nov. 26 two are quite coloured and this group is transferred to 17°—18°, to prevent the high temperature from withering the unfolded flowers too rapidly. On *Dec. 2* 11 of the 12 tulips are in flower; the 12th flower is dried up. With regard to the subjoined description it should be borne in mind, that the groups from hothouse 22°—23° when 1—2 flowers start colouring and those from hothouse $19\frac{1}{2}$°—$20\frac{1}{2}$° when 2—3 flowers begin, are transferred to the hothouse 17°—18°.

From the group exposed to *22°—23° dark* and subsequently to light (Table III *A*) 3 flowers are unfolded, 5 withered, 3 remain white-edged and 1 bulb is diseased on *Dec. 3*. This bad result of this group may be attributed with certainty to the 11 days' darkening, which made the basal part of the stem stretch too much.

Of the group *D, $19\frac{1}{2}$°—$20\frac{1}{2}$° light* (Table III) 10 flowers are in bloom on *Dec. 3*. Whereas from the group which had been kept in the light only 1 flower dried up, 6 flowers withered from the group which had been kept in the dark for some time. To give an impression of the bloom of these early tulips, the one box of the group 17°—19°, next $19\frac{1}{2}$°—$20\frac{1}{2}$° C. light (Table III, group *D*) was photographed on *Dec. 1* (Fig. 6), while Fig. 7 shows the same box on *Dec. 22, 1926*, when the stalks have stretched still further and the colour has grown more intense. As a standard of the lengths the plants attain, it should be mentioned, that from the tip of the bulb to the upperside of the flower the length amounted to 39.0, 36.5, 39.5, 39.0, 37.0 and 35.0 cms. In the groups (Table III, *E* and *F*) which had been in the hothouse 17°—18° the flowers were open on *Dec. 9*; in the group *F*, which was throughout kept in the light, two flowers were withered, in the group *E*, which had partly been kept in the dark, 1 flower. As this group was not so much etiolated, when the covers were removed, it might be expected, that the influence of the dark would not be so conspicuous in this case.

Looking backwards it appears that of the experimental plants which were kept in the light, 31 of the 35 bulbs flowered, while 4 flowers withered;

the groups which had been in the dark for 11 days, did indeed nearly all flower simultaneously with the groups which had only been exposed to the light, but of the 34 tulips only 9 flowered ; 12 withered and 3 had white tips on the tepals.

The temperature-exposure 20°—9°.

From § 3 it already appeared, that the tulips which had been subjected to this preliminary treatment did not attain a nose-length of $4\frac{1}{2}$ cms. (averagely 4.59 cms. $\pm$ 0.12), until Nov. 18, i.e. *a week* later than when exposed to 17°—9°, so that these groups were not distributed over the 4 different hothouses with the temperatures of 22°—23° C., $19\frac{1}{2}$°—$20\frac{1}{2}$° C., and 17°—18° C. before that date (See Tab. IV).

On Nov. 22, when the groups 17°—9° were measured for the sake of comparison the nose-length was also determined in the groups 20°—9°. They cannot yet be distinctly discriminated, it might only be remarked, that the noses of those groups exposed to the higher temperatures, are already slightly longer than those kept in the hothouses 17°—18°. In this short period of time "the keeping in the dark and in the light" has not caused any difference. As in the set 17°—9° we had removed the covers a little late, so that the parts of the stem had grown too vigorously, we now took off the covers of these groups also on this date to prevent them from stretching too much. For mutual comparison figs. 3 and 4 give two boxes on Nov. 23 : both have been exposed to the same hothouse-temperatures and "light", but the tulips of the box in Fig. 3 have been exposed before to 17°—9° : those of Fig. 4 to 20°—9°. The former box was kept in the hothouse for 12 days, the latter for 5 days. Accordingly the relatively inhibitory effect of 20°—9° is evident. When the group 20°—9° has also been in the hothouse for 12 days (Fig. 5), it is, on our comparing this picture with the box in Fig. 3 (the exposure to 17°—9°), equally far advanced as this group was on Nov. 23. The treatment 17°—9° therefore still has a start of 6—7 days.

When the tulips are going to colour in these groups too the boxes are directly transferred to the hothouse 17°—18° C., as it was done with the groups 17°—9°. The non-paraffined boxes which had been kept in a moist hothouse of $19\frac{1}{2}$°—$20\frac{1}{2}$°, were then put in a moist hothouse of 17°—18° C. In these groups too those sets which were kept in 22°—23° C., were the first to flower, viz. group *B* on Dec. 7 and group *A* on Dec. 9 (see table IV) ; from the hothouse $19\frac{1}{2}$°—$20\frac{1}{2}$° group *D* (light and non-paraffined) followed on Dec. 9 and group *C* (dark) on Dec. 13, whilst *E* and *F* from the hothouses 17°—18° also flowered on Dec. 13. From this it follows (see also Table IV), that *keeping dark is not profitable with regard to the period of flowering ;* on the contrary in the higher hothouse-temperatures its influence is rather somewhat *inhibitory* in this respect. Now that we have kept these groups of tulips dark for a shorter period,

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FIG. 6   D 17º 9º 20º LICHT   D 17º 9º20º LICHT   FIG.7
1 DEC.   22 DEC.

they do not suffer from withering or white tips. From the 35 tulips kept in the light in the dry hothouses, 33 came into bloom, 1 withered and 1 showed white tips at the tepals ; in the 36 tulips which had been kept temporarily dark, 33 came into bloom, 2 flowers withered and 1 bore white edges. In the experiments with the preliminary treatment 17°—9° we already noticed, that keeping dark somewhat too long has a detrimental effect due to the stretching of the basal part of the stem, which makes the tulips grow crooked and afterwards in the flowering period causes a strong withering of the flowers or a partial lack of pigmentation, i.e. appearance of white spots. As a period of "dark" does not appear to be a favorable factor for the rapid beautiful flowering of the early tulip *Van der Neer,* the best thing is *to put the tulips in the then desired temperature in the light, when they have attained a nose-length of averagely 4½ cms.*

On our comparing those groups which have been exposed to a preliminary treatment of 17°—9° and 20°—9°, and which have throughout been kept in the light in the hothouses, it appears (Tables III and IV), that *the groups 17°—9° flowered earlier,* viz. respectively those from 22°—23°, 5 days, those from 19½°—20½°, 6 days and those from 17°—18°, 4 days. From which we must conclude that a temperature-exposure to 17° at the beginning of the flower- formation has an *accelerating* effect and choosing this temperature is to be recommended.

It has likewise appeared that these early tulips are not subject to "tipping" ; neither in the dry nor in the moist hothouse did this phenomenon occur in any specimen. The flowers in the moist hothouse 20° were practically as beautiful as in the other hothouses ; from table IV we might be inclined to conclude, that the moist hothouse did not give such fine flowers ; but this must probably be attributed to being kept long in the higher temperature, as at the beginning no moist hothouse 17°—18° was available. Humidity however does not offer any advantages, therefore the temperature of the air and of the boxes need not be kept extra-wet.

In § 1 we called attention to the fact, that for the Darwintulip the temperature-exposure to 17° in summer for early flowering had previously not been chosen, because it would sooner give rise to deviations, and transitions from tepals and leaves might lead to crooked growth. In *Van der Neer,* as far as we could trace, no transitions of different leaves and floral parts occurred neither in 17° nor in 20°. As to the deviation of the normal number of floral parts : 663, this appeared to be by no means greater in 17° than in 20°. In 17° we counted in 27 flowers : 22 of 663 and 5 deviations (viz. 3 with 884, 1 with 763 and 1 with 773) ; on exposure to 20° on the other hand we counted in 39 flowers 30 flowers with 663 and 9 with deviations (viz. 1 with 884, 4 with 773, 3 with 673 and 1 with 774). So it may be, that the various temperatures have an effect on the number of floral parts slightly different from that in the Darwin-tulips, where an increase of the number occurred in the lower temperatures, which difference

was already marked in 23°, 20° and 17°. In a separate research we shall further ascertain the correlation between the various temperatures in summer and the number of floral parts in this "early Tulip".

I wish to conclude with some remarks on the time of planting. Some rests of the group, destined for the control of the stages, were kept dry for some time at a temperature of $9\frac{1}{2}$°—$10\frac{1}{2}$° C. In this way we had got 4 tulips of the preliminary treatment 17°—9° and 5 tulips of that of 20°—9°. Not until *September 6* they were potted up in earthen cylinders and placed at $8\frac{1}{2}$°—$9\frac{1}{2}$°. Simultaneously with the temperature-treatment to which they belonged, they were removed to the hothouse $19\frac{1}{2}$°—$20\frac{1}{2}$°, and now it appeared, that they came into bloom simultaneously with the others in the hothouse $19\frac{1}{2}$°—$20\frac{1}{2}$°. From this it might be concluded that planting later is possible. A subsequent year it will be further investigated whether this might lead to some change in the treatment, which may prove whether the time of planting need be chosen so accurately as we have done now.

§ 5. *Conclusion.*

From the above it has sufficiently appeared, that with a correct treatment the "Early Single Tulip" *Van der Neer* may *flower about December 1—2.* On account of its vigorous flower-stalk this tulip is particularly fit for forcing ; this flower-stalk *does not react upon the light at all,* so that the stalks of all tulips continue as straight as arrows and never show a single curve.

If, therefore, these tulips in the first period of flower-formation (i.e. till Stage III has been reached) are put in 17°, this causes coming into bloom 5—7 days earlier than if they had been put in 20° C. during this first period. If Stage III has been reached, the tulips are transferred to 9° and they are planted after a month (in 9°). When the noses have attained a length of $4\frac{1}{2}$ cms., they may be transferred to the various temperatures in the hothouses. Good care should be taken, that they are put in the light on the tablets, for this is conducive to earlier and finer flowering.

According as higher temperatures are chosen, respectively 22°—23°, $19\frac{1}{2}$°—$20\frac{1}{2}$°, 17°—18°, they will sooner come into bloom ; we should see to it, that in the higher temperatures they are left in these temperatures until they colour, next they should be placed cooler to prevent the flowers from wrinkling. Besides it appeared that the hothouses need not be kept moist to ensure good flowers.

Laboratory for
Plant-physiological Research.

Wageningen, February 1927.

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Physics. — *On the Luminescence produced by bombarding solidified Gases with electric Rays at the Temperature of liquid Helium.* By L. Vegard and W. H. Keesom. Communication N⁰. 186 from the Physical Laboratory at Leiden.

Luminescence from solid Hydrogen.

§ 1. *Aim and Method of the Experiments.* In previous communications[1] results were given of experiments on the luminescence from solid nitrogen, and argon and neon mixed with nitrogen at the temperature of liquid helium.

At the same time it was our object to study the luminescence of solidified hydrogen, but owing to the very low freezing point of this substance, these experiments met with considerable difficulties.

During the experiment a current of hydrogen flows from the discharge tube in which the electric rays are produced, through a boring in a metallic cylinder, through which the rays had to pass, and into the emission chamber, where the gas was condensed on the cold copper surface.

When now the rays were put on, the conditions appeared to be somewhat unstable. The heat transport to the cold surface became so large that a marked increase of temperature resulting in a less effective condensation took place. When the condensation became less effective, the pressure of the observation chamber and the heat transport increased, and in a short time the condensation might cease and the liquid helium would evaporate within 15—20 minutes.

Already in 1924 we were able to examine the luminescence from solid hydrogen produced by swift cathode rays spectroscopically, and it was found to be dominated by continuous bands, and no lines could be seen. The liquid helium, however, did not last sufficiently long for obtaining photographs of the spectrum.

Even with the improved apparatus described in Comm. N⁰. 183 fig. 1, which was used with success for the study of neon-nitrogen luminescence, we were not able to proceed further with solid hydrogen.

The experimental arrangement might be improved in the following way: 1. By increasing the isolation power of the cooling apparatus and by securing good contact between the copper rod and the liquid. 2. By

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[1] L. Vegard, H. Kamerlingh Onnes and W. H. Keesom. These Proceedings **28**, 467; Comm. Leiden N⁰. 173*d*; C. R. **180**, 1084, 1925; Comm. Leiden N⁰. 183, 1926.

diminishing the current of hydrogen necessary for the running of the bulb.

With regard to the last possibility we tried to use a thermionic current — by employing a glowing cathode — but as the heated cathode must be well protected from the cold surface, our attempts in this direction were not successful.

We therefore returned to the ordinary discharge, but tried to reduce the necessary gas current by using a long and narrow boring in the cylinder, which connects the discharge tube with the emission chamber.

In order to increase the isolation power, a new cooling apparatus was made, the construction of which is shown in fig. 1a and b. The apparatus consists entirely of metal and contains three vacuum receivers, one outside the other. The inner receiver (1) contains liquid helium, the second one (2) is to contain liquid hydrogen, and this is surrounded by one (3) which is to be filled with liquid air. Each of the two inner receivers is provided with a tube for filling and one for the evaporated gas and a floater arrangement, which makes it possible to determine the stand of the liquid. The spaces inside and outside the hydrogen receiver (2) communicate and may be evacuated through a side tube. The space outside the liquid air receiver communicates with the emission chamber.

The layer of condensed gas to be bombarded with electric rays is formed on the end of the copper rod (4), which now passes

Fig. 1a.   Fig. 1b.

into the inner receiver, so as to secure an intimate contact with liquid helium. Perhaps the most important point in connection with the cooling
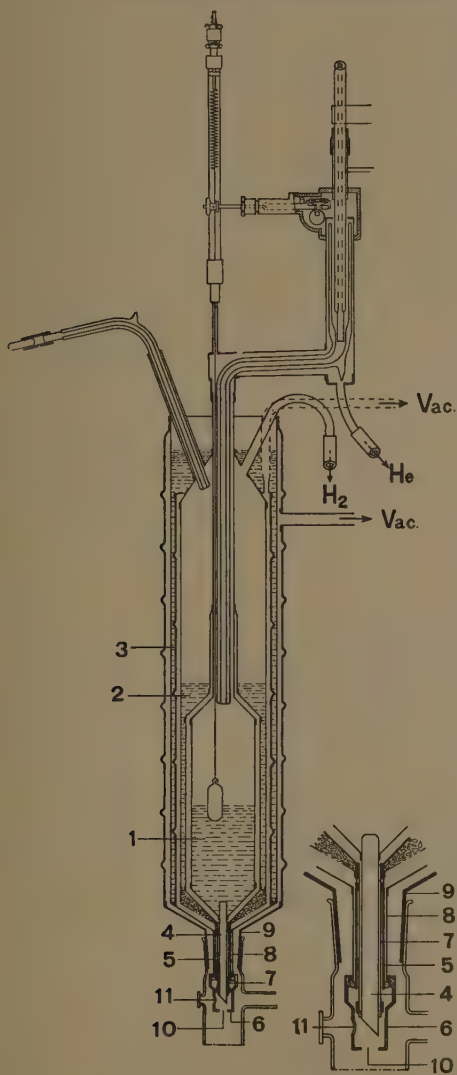
34*

apparatus and that which has caused us the most trouble is the protection of the copper rod (4) from receiving heat from the surroundings.

Already for the apparatus previously described (fig. 1 Comm. N^0 183) a fairly satisfactory construction was found, but for the last apparatus the isolation of the copper rod was considerably improved.

In fig. 1 *b* is shown on a larger scale the construction of the bottom part of the cooling apparatus. The thick brass tube (5) is soldered onto the bottom of the liquid hydrogen receiver (2). A brass cap (6) is screwed onto the lower end of the tube (5). Thus the inner copper rod is surrounded by the tube (5) and cap (6), which during the experiment is maintained at a temperature approximately equal to that of liquid hydrogen.

In order to close the inner vacuum system the brass tube (5) is on the inside connected with the copper rod (4) and on the outside with the liquid air receiver by means of thin tubes of German silver (7) and (8).

The cap (6) is provided with an opening (10) at the bottom for the passage of the rays, and one (11) at the side for the observation of the luminescence.

The discharge tube is fixed to the cooling apparatus by means of a ground joint (9).

By means of this apparatus hydrogen was solidified very effectively on the copper surface, and by a proper choice of the canal that leads to the emission chamber and of the strength of the electric rays a surface of solid hydrogen could be bombarded for several hours. Under favourable circumstances one filling with liquid helium would last for 5—6 hours.

Experimental Results.

§ 2. *Luminescence from solid Hydrogen produced by swift Cathode Rays.* Several experiments have been made to investigate the light emitted when solid hydrogen is bombarded with swift cathode rays produced by induction coil and kenotron rectifier. The rays produced a luminescence of greenish blue colour and of moderate strength. The luminescence was seen to last for at least one minute after the cathode rays were cut off. Consequently a luminescence was produced, which was characteristic of solid hydrogen. In the spectroscope only a continuous spectrum was seen. It might, however, be possible that lines appeared, which were masked by the continuous spectrum. In order to see whether possibly also lines were present, the small spectrograph with high light power was used. The spectrograph was carefully adjusted, so that the continuous spectrum could be seen by means of an auxiliary lens.

Already in the spring 1926 we made in this way an exposure, which lasted for more than one hour. But there was no impression at all to be seen on the plate after development.

Similar experiments have been undertaken at the beginning of this year. In one experiment the exposure lasted for nearly two hours, in another for more than one hour, but with the same negative result.

If we take into account the fact that much less intense sources of light giving a line spectrum would have produced an overexposed spectrogram we can conclude that the line spectrum — if any — emitted from solid hydrogen exposed to swift cathode rays must be extremely faint as compared with the energy emitted in the continuous part.

The absence of any impression on the plate is explained from the continuous character of the spectrum, because the total luminescence, which is of moderate strength is distributed over the whole spectrum.

§ 3. *Luminescence from solid Hydrogen produced by Hydrogen Canal Rays.* Investigations on the luminescence produced when solid hydrogen is bombarded with canal rays were undertaken in June 1926. Bombardment with hydrogen canal rays with velocities corresponding to a parallel sparc gap 3 mm. produced a marked luminescence on the layer of solid hydrogen.

Although the total strength of the luminescence was much smaller than in the case of swift cathode rays, we obtained by means of the same small spectrograph a very good spectrogram by an exposure of about two hours. In both cases we used the same sort of plates (Imp. pancromatic B).

The greater spectrographic effect is due to the fact that the canal rays gave a *line spectrum.*

An enlarged reproduction of the spectrum is given on Fig. 2 N°. 1.
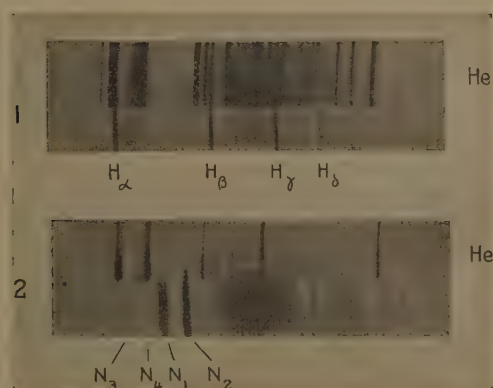
Fig. 2.

As the dispersion is small, the wavelength determinations cannot claim any great accuracy. Still the accuracy is sufficient to show that the very strong lines belong to the Balmer series. In between these lines we notice a number of faint lines.

All lines obtained on the spectrogram (26 in all) are given in the

table. The accuracy of measurement in the case of isolated strong lines may be estimated by means of the values found for the lines of the BALMER series, the identification of which is certain. In the case of faint

TABLE I.

N⁰.	Solid hydrogen		Gaseous hydrogen	
	λ	I	λ	Designation
1	6559.0	100	6563.0	H_α
2	6324.9	5	6327.1	$3\alpha3$ Q (1)
3	6214.5	5	6224.8	$2\alpha2$ Q (1)
4	6106.8	10	6121.8	$1\alpha1$ Q (1)
5	6018.6	2	6018 3	$0\alpha0$ Q (1)
6	(5952.7)	2 (?)	5950.2	
7	(5796.0)	1 (?)	5796.8	
8	4861.7	80	4861.6	H_β
9	4680.5	10	4680.4	$3\beta3$ Q (1)
10	4629.5	20	4617.5	$2\beta2$ Q (1)
11	4578.5	5	4577	$1\beta1$ Q (m) (?)
12	4557.6	5	4554	$1\beta1$ Q (1)
13	4490.8	10	4490.6	$0\beta0$ Q (1)
14	4453.1	10	4453.2	
15	4418.0	5	4417.5	
16	4340.3	80	4340.8	H_γ
17	4308.8	10	4308.6	$4\beta3$ Q (1)
18	4204.0	10	4205.3	
19	4179.3	10	4182.1	$3\gamma3$ Q (1)
20	4161.0	5	4159.3	$3\beta2$ Q (1)
21	4101.5	35	4101.9	H_δ
22	4085.2	2	4085.2	$\{ 1\beta0$ Q (1) $\atop 1\gamma1$ Q (3)
23	4068.0	15	4069.8 (67.1)	
24	4025.8	5	4024.8	$0\gamma0$ Q (1)
25	3995.3	5	3995.3	
26	3970.8	5	3970.8	H_γ

lines and lines not well separated the error may be considerably greater than in the case of the BALMER lines.

In the third column is given the relative intensities of the lines as they were roughly estimated from the spectrogram.

With regard to the fairly small number of faint lines appearing in between the strong BALMERlines, they might either be lines characteristic of the solid state or a certain selection of lines belonging to the ordinary second hydrogen spectrum.

In the latter case we should on account of the extremely low temperature expect to find either rotational zero lines or lines corresponding to a small rotational quantnumber.

Among the lines of the second hydrogen spectrum we find actually lines, which within the limit of possible errors coincide with those observed from solid hydrogen.

Although the second spectrum of hydrogen has a large number of lines, we cannot assume all coincidences to be accidental, and the correctness of the interpretation is greatly strengthened when we consider the series symbols of the lines appearing.

A great number of the lines of the second hydrogen spectrum has now been classified into band series and interpreted in terms of the general theory of band spectra. A summary of the present state of these investigations has been given by RICHARDSON [1]).

From the theory of band spectra we know that the frequency depends on the following conditions:

1. An electron jump between stationary orbits. For the class of lines here considered RICHARDSON finds that the electron jumps correspond to those of the BALMER-series, which take place towards a stationary orbit corresponding to a principal quantnumber 2. These electronic jumps are denoted by α, β, γ, corresponding to $H_\alpha, H_\beta, H_\gamma$ of the BALMERseries.

2. The number of oscillatory quanta possessed by the system at the beginning and end of the emission process. The oscillatory quantnumber at the start RICHARDSON denotes by s, and that at the end by r.

3. The rotational energy at the beginning and end of the emission process. The rotational quantnumber (m) cannot change by more than one unity.

With the denomination introduced by RICHARDSON lines of the second hydrogen spectrum are indicated by symbols of the form:

$$r \, \alpha \, s \, Q \, (m) \text{ or}$$
$$r \, \beta \, s \, Q \, (m).$$

The interpretation of the observed lines in terms of the symbols of RICHARDSON is given in the last column of the table.

[1]) O. W. RICHARDSON, Proc. Roy. Soc. **113**, 368, 1926.

II

§ 4. *Luminescence from solid Nitrogen produced by Canal Rays at liquid Helium Temperature*. In previous communications one of us [1] has described results of experiments undertaken at the temperature of liquid hydrogen, where solidified gases have been exposed to canal rays. In the case of solid nitrogen it was found that the luminescence consisted of the four bands $N_1 N_2 N_3 N_4$, a diffuse band series (A), and some other lines and bands.

Similar experiments have been made at the temperature of liquid Helium.

In order to see whether the diffuse (A) bands were emitted at this very low temperature, we used the small glass spectrograph. A reproduction of the spectrum obtained is shown in N⁰ 2 of Fig. 2. The position of the lines and bands will appear from table II.

It appears that the four bands $N_1 N_2 N_3 N_4$ are emitted. The series A also appears, but with an intensity distribution of the bands different from that found at the temperature of liquid hydrogen. In the case of liquid helium there is one maximum at 4610 of great intensity, but the intensity of the other bands is relatively much smaller than in the case of liquid hydrogen.

Some of the other lines and bands given in II have also been observed at the temperature of liquid hydrogen, but we also notice some new lines such as 6004, 5766, 3870, 3849. As to the nature of these lines,

TABLE II.

6379	N_3
6004.2	
5944.9	N_4
5765.9	
5628.0 ⎱	N_1
5548.5 ⎰	
5230.3	N_2
4790 max.	A_6
4610 max.	A_5
4315	
4255 max.	A_4
(4169.1) diffuse	
4057.4 2. *P.B.* I	
3996.1 „ „ II	
3881.3 *N.B.* II	
3870.1	
3849.3	
3802.5 2. *P.B.* I	
3750.7	

some of them seem to belong to the positive band spectrum of nitrogen. These are indicated in the table with the series symbols used in H. KAYSER, „Handbuch der Spektroskopie".

[1] L. VEGARD, Comm. Leiden N⁰. 175; Det Norske Vid. Akad. Skr: 1925, N⁰. 9; Ann. d. Phys. (4) **79**, 377, 1926.

Chemistry. — *Equilibria in systems, in which phases, separated by a semi-permeable membrane.* XXI. By F. A. H. SCHREINEMAKERS.

Influence of the pressure on osmotic systems and on the osmotic pressure.

In the previous communication we have deduced some properties (a.o. the change of the osmotic pressure of a system E at change of the pressure of the water) with the aid of the $P_0\pi$-diagram. Of the several other diagrams, which we can use for this also, we shall take now the $P\xi$-diagram; consequently we draw on the one axis the pressure P and on the other axis the ξ viz. the $O.W.A.$ of a system.

If we take f.i. in fig. 1 the point d, then this represents a system which has a pressure $P(d) = Ou$ and an osmotic water-attraction $\xi(d) = ud$. [Of course the length of the line ud does not represent the absolute value of $\xi(d)$, but this value, increased or decreased with an amount, which is the same for all systems in the same diagram. By this the difference between two ξ's f.i. between $\xi(d)$ and $\xi(n)$ or between $\xi(d)$ and $\xi(k)$ does not change; consequently this has no influence on our considerations, in which we only use the difference of two $O.W.A.$'s].

If we represent the $O.W.A.$ and the molecular volume of water by ξ_0 and v_0, then we have:

$$d\xi_0 = -v_0 \cdot dP \quad \ldots \ldots \ldots \ldots \quad (1)$$

so that the $O.W.A.$ of the water decreases on increase of pressure. If we draw on the P-axis the pressure and on the ξ-axis the corresponding $O.W.A.$ of the water, then we get, therefore, a $P\xi$-curve like mr in fig. 1. As v_0 decreases at increase of pressure, but as this decrease is very small only, mr is a curve convex to the abscis, which is a right line at approximation.

We now assume that the temperature, for which fig. 1 is valid, is higher than the melting-point of ice under its own vapour-pressure. Under a definite pressure $P(m)$ the water then passes into:

$$water + water\text{-}vapour. \quad \ldots \ldots \ldots \quad (2)$$

which passes into water-vapour on further decrease of pressure. The $P\xi$-curve of the water terminates on decrease of pressure, therefore, in the point m (fig. 1) where the $P\xi$-curve ma of the water-vapour begins. This is defined in each point by:

$$d\xi_0 = -V_0 \cdot dP \quad \ldots \ldots \ldots \ldots \quad (3)$$

in which ξ_0 and V_0 represent the $O.W.A.$ and the molecular volume of the water-vapour. As $V_0 > \dot{v}_0$ the two curves are situated in the point m, with respect to one another, as is drawn in the figure. This

Fig. 1.

curve ma, of which we imagine the point a at infinite distance, is convex to the abscis and approaches asymptotically the ξ-axis; the $O.W.A.$ of the water-vapour increases continuously on decrease of pressure, in order to become infinitely large under the extremely small pressure $P(a)$.

The $P\xi$-curve of the water consists, therefore, of the two branches mr and ma, the first of which indicates the $O.W.A.$ of the liquid water and the second that of the water-vapour; the point m represents the $O.W.A.$ of the system (2) occurring under the pressure $P(m)$.

If we compress, starting from the pressure $P(a)$, water-vapour to water of the pressure $P(r)$ then we pass along the $P\xi$-curve from a towards r, so that the $O.W.A.$ decreases from $\xi(a)$ till $\xi(r)$. This change of the $O.W.A.$ is continuous along the branches, in the point m, however, it is discontinuous.

We now take a binary system E, consisting of the components: $Y + water$. We assume that Y is a solid substance with such a small vapour-pressure, that it does not occur in the vapour under the considered pressures. Under a very small pressure we then have the system:

$$E_1 = Y + water\text{-}vapour \quad . \quad . \quad . \quad . \quad . \quad (4)$$

in which Y occurs as solid substance. If we raise the pressure, then this system passes under a definite pressure $P(c)$ which is smaller than $P(m)$ into the system:

$$E_2 = (Y + L + water\text{-}vapour) \quad . \quad . \quad . \quad . \quad . \quad (5)$$

As this system has no freedoms (2 components in 3 phases, while the

temperature is constant) it exists under a definite pressure $P(c)$ only and the liquid L has a definite composition. At further increase of pressure is formed a system:

$$E_3 = Y + L \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

We now shall assume that the solubility of the solid substance Y increases at increase of pressure. With increase of pressure then is formed under a definite pressure $P(s)$ the system:

$$E_4 = L \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

in which the liquid L has a definite composition and which remains on further increase of pressure. (We assume viz. that no new phase will be formed on further increase of pressure, as f. i. a hydrate of Y or a second liquid).

While the pressure $P(c)$ under which E_1 passes into E_2 and E_2 into E_3, is independent on the ratio of the quantities Y and water, this is not the case with the pressure $P(s)$. Viz. $P(s)$ is larger, according as the system E_1 contains more solid Y and less water-vapour.

Consequently the system E occurs, according to its pressure, in several states, viz. as:

$$
\left.
\begin{array}{lll}
E_1 = Y + \textit{water-vapour} & \textit{when } P \leqq P(c) & \\
E_2 = Y + L + \textit{water-vapour} & \textit{when } P = P(c) & \\
E_3 = Y + L & \textit{when } P \gtrless P(c) \text{ and } P < P(s) & \\
E_4 = L & \textit{when } P \geqq P(s) &
\end{array}
\right\} \quad (8)
$$

As the system E_1 contains water-vapour, its $O.W.A.$ is the same as that of water-vapour only; the $P\xi$-curve of system E_1 coincides, therefore, with curve am of fig. 1. As the highest pressure $P(c)$ under which system E_1 still consists, is lower than the pressure $P(m)$, this curve does not continue till in m, but it terminates already in c. Consequently the $P\xi$-curve of the system E_1 consists of curve ac, the final-point c of which represents also the $O.W.A.$ of the system E_2.

In this point c also begins the $P\xi$-curve of the system E_3, the direction of which is defined in every point by:

$$(d\xi)_3 = - \triangle V_E . dP \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

If we represent the molecular volume of the solid substance Y and of the liquid L by V_y and V, then we have [compare formula's (10^a) — (16) of Communication XIX]:

$$\triangle V_E = \frac{V - yV_y}{1 - y} = \triangle V_W + \frac{y}{1-y} . \triangle V_y \quad . \quad . \quad (10)$$

wherein for the sake of abbreviation:

$$\triangle V_W = V - y \frac{\partial V}{\partial y} \qquad \triangle V_y = V + (1-y) \frac{\partial V}{\partial y} - V_y \quad . \quad (11)$$

Now we shall show that the $P\xi$-curve of system E_3 is represented by a curve cfl (fig. 1) which has a minimum in f and, if we represent

the melting-pressure of the substance Y by $P = Ov$, approaches asymptotically the vertical line vv'; we imagine, therefore, the point l of this curve, on infinite distance.

We have assumed viz. that the solubility of the substance Y increases with increase of pressure; $\triangle V_y$ is negative, therefore [this follows f. i. from formula (15) of Comm. XIX]. Just as previously we shall take $\triangle V_w$ positive. It now appears from (10) that $\triangle V_E$ is certainly negative for values of y which differ no much from the unity, consequently under pressures which differ no much from the melting-pressure $P = Ov$ of the substance Y. Under lower pressure $\triangle V_E$ can be as well positive as negative; if, as we shall assume, Y is sufficiently small under low pressures, then $\triangle V_E$ is positive, therefore.

It follows from (9) that $(\delta\xi)_3$ is negative under lower pressures and positive under higher pressures. Starting from point c the $P\xi$-curve must, therefore, firstly fall till in a point f, where the $O.W.A.$ is a minimum and it must rise afterwards, in order to approach the line vv' assymptotically.

The branches ac and cf form an angle with one another in the point c; this follows from (3) and (9), the first of which defines the direction of ac and the second that of clf in the point c. [Of course we have to give to V_0 and $\triangle V_E$ the values, which they have in the point c, consequently under the pressure $P(c)$].

Above we have seen that system E_3 passes into system $E_4 = L$ under a pressure $P(s)$ and that $P(s)$ is larger, according as system E_1 contains more solid Y and less water-vapour.

We now imagine this point s anywhere on curve cfl f. i. in the point g; the $P\xi$-curve of the system $E_4 = L$, starting from this point is then defined by:

$$(d\xi)_4 = - \triangle V_W \cdot dP \quad . \quad . \quad . \quad . \quad . \quad . \quad (12)$$

of which the value of $\triangle V_W$ is given by (11). As we take $\triangle V_W$ positive and consequently the $O.W.A.$ of the system decreases with increase of pressure, we may represent the $P\xi$-curve, therefore, by the dotted curve gg'. If we consider, as is done in fig. 1, stable states, only, then this curve goes, starting from g only towards higher and not towards lower pressures; the system $E_4 = L$ is viz. metastable under pressures smaller than $P(g)$ and it is converted into $E_3 = Y + L$.

As point s can coincide with every arbitrary point of curve cfl, from each point of cfl a similar curve is starting, three of which, viz. ee', gg' and hh' are drawn.

It is clear, without more, that the curves gg' and hh' are situated below the curve cfl; it follows from (9) and (12) that this is also the case for ee'. If we consider viz. the value of $\triangle V_E$ from (10) then follows:

$$(d\xi)_3 - (d\xi)_4 = - \frac{y}{1-y} \cdot \triangle V_y \cdot dP \quad . \quad . \quad . \quad . \quad (13)$$

As $\triangle V_y$ is negative in every point of curve cfl, follows for positive values of dP:

$$(d\xi)_3 > (d\xi)_4 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

from which the above mentioned follows. Of course the same is true for the curve, starting from point c, which we have to imagine below cfl.

We are able to deduce the above still also in the following way. For this we use the rule, deduced in Communication XVI, that the $O.W.A.$ of a system (at constant T and P) becomes smaller, when taking water in.

We imagine viz. in fig. 1 to be drawn a straight line, which intersects curve ef in a point x and ee' in a point x'. Point x represents a system $Y + L$ and x' a liquid L; both systems have the same pressure. As the liquid L' must have a greater amount of water than the system $Y+ L$, consequently L' can arise from $Y+ L$ by taking water in; therefore, L' must have also a smaller $O.W.A.$ than $Y + L$. Consequently the point x' must be situated in fig. 1 below point x and, therefore, curve ee' below curve ef.

If we compress, starting from the low pressure $P(a)$, the system E_1, then this passes under the pressure $P(c)$ firstly into E_2 and afterwards into E_3, which is converted into $E_4 = L$ under a pressure $P(s)$.

If we take $P(s) = P(g)$ then the system goes along the curves $a\,c$, cfg and gg'; consequently its $O.W.A.$ firstly decreases from a till in f, where it becomes a minimum, then it increases till in g, in order to decrease again at further increase of pressure.

Of the many osmotic equilibria, which we can deduce from fig. 1, we only shall indicate some. If we draw in fig. 1 a horizontal line, then its points of intersection with the $P\xi$-curve represent different states of a system E which have the same $O.W.A.$ and may be, therefore, in osmotic equilibrium with one another. We have f.i. the osmotic equilibria:

$$(Y+water\text{-}vapour)_b \mid (Y + L)_k . \quad . \quad . \quad . \quad . \quad (15)$$

$$(Y + L + water\text{-}vapour)_c \mid (Y + L)_l . \quad . \quad . \quad . \quad . \quad (16)$$

in which the system at the left side of the membrane has a low pressure and the other a high pressure. Further we have f.i. still the osmotic equilibria:

$$(Y + L)_d \mid (Y + L)_h$$
$$(Y + L)_e \mid (Y + L)_g \mid (Y + L)_{h'}$$

in which also the pressure at the left side of a membrane is smaller than at the right side.

With the aid of fig. 1 we can also deduce the osmotic pressure of a system and the change which this endures, if we change the pressure of the water. We then get the $P_0\,\pi$-diagram ($P_0 =$ the pressure of the

water and $\pi =$ the osmotic pressure of the system) discussed in the previous communication. Here however, we shall mean with osmotic pressure not only the difference in pressure on both sides of the membrane, if water is at one of the sides, but also if there occurs water-vapour instead of water.

As is apparent from fig. 1 we can have f.i. the osmotic equilibrium:

$$(water)_n \mid (Y + L)_e \qquad \pi = P(e) - P(n) = en \quad . \quad . \quad . \quad (17)$$

in which at the left side of the membrane is a pressure $P(n)$ and at the right side a pressure $P(e)$. As, therefore, the osmotic pressure of this system is $\pi = P(e) - P(n)$ this is represented in fig. 1 by the line en.

We can represent system (17) in the $P_0\pi$-diagram (fig. 2) by a point e; the pressure $P(n)$ of the water is then indicated by On and the osmotic pressure π by en (equal to en in fig. 1).

It appears from fig. 1 that water under the pressure $P(n)$ yet also can be in osmotic equilibrium with the system $Y + L$ under the pressure $P(g)$. Consequently we also have the osmotic equilibrium:

$$(water)_n \mid (Y + L)_g \qquad \pi = P(g) - P(n) = gn \quad . \quad . \quad . \quad (18)$$

The pressure $P_0 = P(n)$ of the water and the osmotic pressure $\pi = gn$ of the system are represented, therefore, in fig. 2 by the point g.

If we take f.i. the osmotic equilibria:

$$(water + water\text{-}vapour)_m \mid (Y + L)_d \qquad \pi = P(d) - P(m) = dm \quad . \quad (19)$$

$$(water + water\text{-}vapour)_m \mid (Y + L)_h \qquad \pi = P(h) - P(m) = hm \quad , \quad (20)$$

from fig. 1, then we can represent them in fig. 2 by the points d and h.

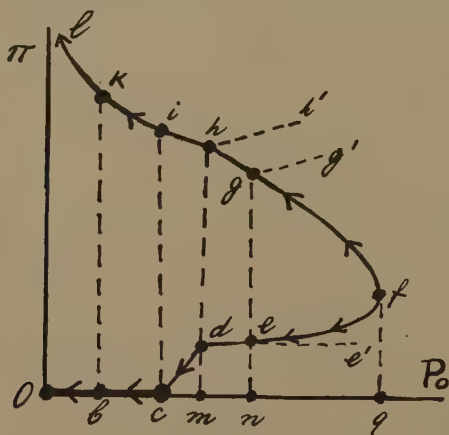If we do in corresponding way with the other osmotic equilibria of

Fig. 2.

fig. 1, then we find a $P_0\pi$-diagram as in fig. 2; the corresponding points in those figures are indicated by the same letters.

We now take from fig. 1 the osmotic equilibrium:

$$(water-vapour)_b \mid (Y + water-vapour)_b \qquad \pi = 0 \quad . \quad . \quad . \quad (21)$$

in which the pressure must be the same on both sides of the membrane; as, therefore, the osmotic pressure is zero, consequently it is represented in fig. 2 by the point b situated on the P_0-axis.

If we heighten the pressure at the one side of the membrane, then we must heighten the pressure at the other side with a same amount till under a definite pressure $P(c)$ the osmotic equilibrium:

$$(water-vapour)_c \mid (Y + L + water-vapour)_c \qquad \pi = 0 \quad . \quad . \quad (22)$$

occurs. Consequently the equilibrium goes in fig. 1 along the curve bc and in fig. 2 along the straight line bc.

We now imagine in fig. 1 a horizontal line, which intersects cm in a point m' and cd in a point d'. We then have the osmotic equilibrium:

$$(water-vapour)_{m'} \mid (Y + L)_{d'} \qquad \pi = d'm' \quad . \quad . \quad . \quad (23)$$

in which the pressure $P(d')$ at the right side of the membrane is larger than the pressure $P(m')$ at the left side. Consequently the system has, as is indicated in (23) an osmotic pressure $\pi = d'm'$; the point d' (not drawn) is situated, therefore, in the $P_0 \pi$-diagram no more on the P_0-axis, but above it. If in fig. 1 m' goes along curve cm (and, therefore, d' along curve cd), then therefore in fig. 2 m' goes along the line cm and d' along the curve cd.

If we bring the pressure at the left side of the membrane to $P(m)$, then we have to bring the pressure at the right side of the membrane to $P(d)$; we then get the osmotic equilibrium (19) with its osmotic pressure $\pi = dm$.

If we raise the pressure at the left side of the membrane, still further f.i. to $P(n)$, then we must raise the pressure at the other side to $P(e)$; we then get the osmotic equilibrium (17) with the osmotic pressure $\pi = en$.

With still further increase of pressure, arises the osmotic equilibrium:

$$(water)_q \mid (Y + L)_f \qquad \pi = fq \quad . \quad . \quad . \quad . \quad (24)$$

represented in the two diagrams by f and q.

If we raise the pressure of the water till above $P(q)$ f.i. to $P(r)$ then, independent which pressure we make at the right side of the membrane, no more an osmotic equilibrium:

$$(water)_{P(r)} \mid (Y + L)_P \qquad \pi = P - P(r) \quad . \quad . \quad . \quad (25)$$

can exist; the difference in pressure $P - P(r)$ is, therefore, no osmotic pressure. It appears viz. from the $P\xi$-diagram (fig. 1) that the $O.W.A.$ of the water under the pressure $P(r)$ is smaller than that of the system $Y + L$ under every arbitrary pressure P; consequently water must diffuse from left to right. Therefore, we can raise the pressure of the water till

a definite pressure $P(q)$ only; therefore, above this pressure the osmotic equilibria, discussed above, can exist no more.

As the system $Y + L$ goes along curve fl of fig. 1 with further increase of pressure, with which its $O.W.A.$ increases, we must lower the pressure of the water, therefore. We then get f.i. the osmotic equilibrium (18) with the osmotic pressure $\pi = gn$. The ascending branch fl in fig. 1 becomes, therefore, in fig. 2 a branch, descending from point f.

If we raise the pressure at the right side of the membrane to $P(h)$, then it must decrease at the left to $P(m)$; we then get the osmotic equilibrium (20) with the osmotic pressure $\pi = hm$.

If we raise still further the pressure at the right side of the membrane, then it must fall again at the left side of the membrane, so that the water passes into vapour. We then get f.i. the osmotic equilibria:

$$(watervapour)_c \mid (Y + L)_i \qquad \pi = ic \quad . \quad . \quad . \quad . \quad (26)$$

$$(watervapour)_b \mid (Y + L)_k \qquad \pi = kb \quad . \quad . \quad . \quad (27)$$

in which at the left side of the membrane is a very small pressure, but at the right side a very large pressure, so that the osmotic pressure also is very large. Both systems are indicated in the $P_0\pi$ diagram by the points i and k.

The $P_0\pi$ curve of the system consists, therefore, of four parts viz. of the straight line Oc and the three curves cd, dfh and hl. With the aid of fig. 1 we find that the $O.W.A.$ of the system must increase along the $P_0\pi$-curve of fig. 2 in the direction of the arrows.

The dotted curves ee', gg' and hh' of fig. 1 indicate, as we have seen above, the $O.W.A.$ of systems $E_4 = L$; if we represent also their osmotic pressures in fig. 2, then we get herein the homonymous and also dotted curves; starting from their beginning-points e, g and h they ascend or descend a little.

If we compare the $P_0\xi$-diagram of this communication with that of the previous, which is deduced in a total other way then we see that both, although they correspond in substance, yet show difference. The diagram of the previous communication is viz. deduced for an entirely arbitrary system; however, fig. 2 of this communication is valid for a special case viz. if the system passes successively the states, indicated in (8).

We now take the osmotic system:

$$\boxed{} \overset{\longleftarrow}{\underset{|}{}} (Y + L)_k \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (28)$$

in which at the left side of the membrane a closed empty space, the volume of which we keep constant; at the right side is the system $Y + L$ under the constant pressure $P(k)$ represented by point k of fig. 1. Without more it is clear that water will diffuse towards the left; of course we can also say that the $O.W.A.$ at the left of the membrane (in fig. 1 represented by point a) is very large with respect to the

$O.W.A. = \xi(k)$ which is on the right. According as more water-vapour comes, however, in the closed space, the tension of this vapour increases and its $O.W.A.$ decreases, therefore; if this tension has increased to $P(b)$ and consequently the $O.W.A.$ has decreased to $\xi(b) = \xi(k)$, then the diffusion of the water-vapour stops and the osmotic equilibrium:

$$\boxed{\underset{b}{water\text{-}vapour}} \; | \; (Y + L)_k \qquad \pi = k\,b \; . \; . \; . \; . \; (29)$$

with the osmotic pressure $\pi = k\,b$ is formed.

We now take instead of (28) the osmotic system:

$$\boxed{} \leftarrow\!\!|\; (Y + L)_g \; . \; .\,. \; . \; . \; . \; . \; (30)$$

which has on the right a pressure $P(g)$ and an $O.W.A. = \xi(g)$ (fig. 1). If so much water-vapour has diffused towards the left, that the closed space is saturated with water-vapour, then is at the leftside of the membrane the pressure $P(m)$ and the $O.W.A. = \xi(m)$; as, however, $\xi(m)$ is still greater than $\xi(g)$, the whole space must be filled with liquid water, the pressure of which will rise till its $O.W.A.$ is fallen to $\xi(g)$. It appears from fig. 1 that this is the case, if the water gets a pressure $= P(n)$ and an $O.W.A. = \xi(n) = \xi(g)$. (30) now passes into the osmotic equilibrium:

$$\boxed{\underset{n}{water}} \; | \; (Y + L)_g \qquad \pi = g\,n \; . \; . \; . \; . \; . \; (31)$$

If we take, therefore, an osmotic system:

$$\boxed{} \leftarrow\!\!|\; (Y + L)_x \; . \; . \; . \; . \; . \; . \; . \; (32)$$

then it depends on the pressure $P(x)$ at the right side of the membrane, which of the two osmotic equilibria:

$$\boxed{\underset{x'}{water\text{-}vapour}} \; | \; (Y + L)_x \qquad \boxed{\underset{x'}{water}} \; | \; (Y + L)_x \; . \; . \; (33)$$

will be formed from this. The first is formed with very small and also with very large values of $P(x)$, viz., as is apparent from fig. 1, if $P(x)$ is smaller than $P(d)$ or larger than $P(h)$. The second arises if we take $P(x)$ between $P(d)$ ond $P(h)$.

If we bring into the closed space the solid substance Y, so that we have the osmotic system:

$$\boxed{Y} \leftarrow\!\!|\; (Y + L)_x \; . \; . \; . \; . \; . \; . \; . \; (34)$$

then this passes into one of the osmotic equilibria

$$\boxed{\underset{x'}{Y + \; water\text{-}vapour}} \; | \; (Y + L)_x \qquad \boxed{\underset{x'}{Y + L}} \; | \; (Y + L)_x \; . \; . \; (35)$$

The first is formed only with very high values of $P(x)$ viz. if $P(x)$ is greater than $P(i)$; the second arises when $P(x)$ is situated between $P(c)$ and $P(i)$.

With the second of the equilibria (35) we can distinguish two cases. The point x' viz. is always situated on branch cf while x can be situated as well on cf as on fi. If x is situated on cf then is formed, therefore, at the left of the membrane the same system as is present at the right side and $P(x')$ becomes $= P(x)$. If, however, x is situated on fi, then both systems are different and $P(x') < P(x)$.

We now imagine the closed space to be filled with water-vapour, we put the pressure $= P_0$, the thermodynamical potential of the water-vapour $= C_0$ and its $O.W.A. = \xi_0$. We then have:

$$\xi_0 = - C_0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (36)$$

We bring into this space still $q \, Mol \, X + r \, Mol \, Y \ldots$, in which X and Y are substances which pass into vapour, f.i. alcohol, aceton etc. If there were already n Mol. water-vapour, then, if this vapour follows the gas-laws, in this space the pressure will rise to

$$P = \frac{n + q + r \ldots}{n} P_0 \quad . \quad . \quad . \quad . \quad . \quad (37)$$

This vapour G has then a composition:

$$x \, Mol \, X + y \, Mol \, Y + \ldots + (1 - x - y \ldots) \, Mol \, W \quad . \quad . \quad (38)$$

in which $x, y \ldots$ have a definite value and

$$1 - x - y \ldots = \frac{n}{n + q + r \ldots} \quad . \quad . \quad . \quad . \quad (39)$$

The addition of those new substance will enlarge the $O.W.A.$ of the vapour, if we keep the pressure $= P_0$; the increase of the pressure from P_0 to P causes, however, the falling of this $O.W.A.$; we now shall show that both influences compensate one another, so that the $O.W.A.$ remains unchanged.

If we take the vapour G under a pressure P_0 then its $O.W.A.$ is defined by:

$$\xi (P_0) = - \zeta + x \frac{\partial \zeta}{\partial x} + y \frac{\partial \zeta}{\partial y} \ldots \quad . \quad . \quad . \quad . \quad (40)$$

In accordance with formula (4ª) of communication XVII we can write for this:

$$\xi (P_0) = - C_0 - RT \, log \, (1 - x - y \ldots) \quad . \quad . \quad . \quad (41)$$

As $log \, (1 - x - y \ldots)$ is negative, it appears from this and from (36) that the $O.W.A.$ increases, if we keep the pressure P_0. We now bring the pressure from P_0 to P. If we represent the molecular volume of the water-vapour by V_0, then we have:

$$d\xi = - V_0 . dP \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (42)$$

Then follows for the $O.W.A.$ of the vapour G under the pressure P_1

$$\xi = \xi(P_0) - \int_{P_0}^{P} V_0 \, . \, dP = \xi(P_0) - RT \log \frac{P}{P_0} \quad . \quad . \quad . \quad (43)$$

so that we may write with the aid of (41)

$$\xi = -C_0 - RT \log(1 - x - y \ldots) - RT \log \frac{P}{P_0} \quad . \quad . \quad . \quad (44)$$

If we substitute in this the values of $1 - x - y$ from (39) and that of $P : P_0$ from (37) then follows:

$$\xi = -C_0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (45)$$

It appears, therefore, from (36) and (45) that the $O.W.A.$ has not changed. Consequently it follows from this for a vapour which follows the gas-laws:

the $O.W.A.$ of a vapour, confined in a space of constant volume, does not change, if we add to this or withdraw from this one or more substances (excepted water).

Above we have seen that the osmotic system (28) passes into (29) with the occurrence of the state of equilibrium. We now fill the closed space of (28) with a gas or mixture of gases of a pressure $P(z)$ which does not contain water-vapour, (f. i. air, alcohol, aceton etc.). We then have the osmotic system:

$$\boxed{gas} \underset{z}{\overset{|}{\longleftarrow}} (Y + L)_k \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (46)$$

in which watervapour will diffuse towards the left till is formed the osmotic equilibrium:

$$\boxed{gas} \underset{z'}{\overset{|}{}} (Y + L)_k \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (47)$$
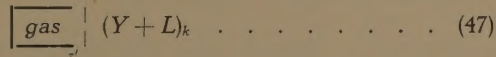
With the transition of (28) into (29) the pressure at the left side of the membrane rises from zero to $P(b)$; it now follows from our previous considerations that the pressure must increase with the same amount at the transition of (46) into (47); consequently we have: $P(z') = P(z) + P(b)$. The quantity of water-vapour, which diffuses towards the left, is, therefore dependent only on the volume of the closed space, independent whether this was firstly vacuum or filled with a mixture of gases.

(*To be continued.*)